# Self-pulsing in a band model for dye lasers

Hong Fu and H. Haken

Institut für Theoretische Physik und Synergetik, Universität Stuttgart, 7000 Stuttgart 80, Federal Republic of Germany

(Received 2 February 1990)

We study the self-pulsing stemming from the Risken-Nummedal-Graham-Haken-type multimode instability in the dye laser described by a band model. Analytical self-pulsing solutions for arbitrary pumping are presented. A distinct feature of the pulsation is that it corresponds to a very low pumping threshold and the required cavity-mode condition can be satisfied in a conventional ring dye laser. Another distinct feature is that the phase velocity of the pulsation may be smaller than the light velocity in the medium. The simple rule, which tells us whether the pulsation is a super- or subcritical one and was found for the two-level model in a previous paper, has been extended to the band model. The results are relevant for experimental investigation of the Risken-Nummedal-Graham-Haken-type multimode instabilities, which are intrinsic to multimode lasers, but have not yet been identified in experiment.

### I. INTRODUCTION

The present paper deals with the self-pulsing developing from the Risken-Nummedal-Graham-Haken (RNGH) -type multimode instabilities in ring dye lasers. It is a succession of our previous work on multimode instabilities in the band model for dye lasers<sup>1</sup> and an extension of the analytical self-pulsing solutions obtained for two-level systems.<sup>2</sup>

## A. RNGH instability in two-level systems

The multimode instability in a homogeneously broadened unidirectional ring laser was first investigated by Risken and Nummedal<sup>3</sup> and Graham and Haken<sup>4</sup> in 1968. They found that a cavity mode propagating in the z direction with infinitesimal amplitude A in the form

$$A \exp(i\alpha_{\nu} z), \quad \alpha_{\nu} \equiv \frac{2\pi\nu}{L} \quad (\nu = 0, \pm 1, \pm 2, ...), \quad (1.1)$$

where L is the cavity length, grows to destroy the stability of the stationary solution if the wave number  $\alpha_v$  falls into the regime

$$\alpha_{\min}(\Lambda) \le |\alpha_{\nu}| \le \alpha_{\max}(\Lambda) , \qquad (1.2)$$

where  $\Lambda$  is the dimensionless pump parameter. In the  $(\Lambda, \alpha)$  plane the upper and lower boundaries of the instability region are given by

$$\alpha_{\max,\min} = \frac{\gamma_{\perp}}{c} \left[ \frac{\gamma}{2} (3\Lambda - \gamma \pm R) \right]^{1/2} \left[ 1 - \frac{2\chi}{\Lambda - 2 - \gamma \pm R} \right],$$
(1.3)

with  $R \equiv [\Lambda^2 - 2(4 + 3\gamma)\Lambda + \gamma^2]^{1/2}$ ;  $\gamma \equiv \gamma_{\parallel}/\gamma_{\perp}$  and  $\chi \equiv \kappa/\gamma_{\perp}$ , where  $\kappa$ ,  $\gamma_{\perp}$ , and  $\gamma_{\parallel}$  are the relaxation constants of the electric field, the polarization, and the inversion respectively; *c* is the light velocity in the medium. Mean-

ingful solutions  $\alpha_{\min}(\Lambda)$  and  $\alpha_{\max}(\Lambda)$  exist only for

$$\Lambda \ge \Lambda_{c,\min} = 4 + 3\gamma + 2\sqrt{2(1+\gamma)(2+\gamma)} .$$
 (1.4)

By increasing  $\Lambda$  the instability condition (1.2) is satisfied by fulfilling either

$$\alpha_{\nu_c} = \alpha_{\max}(\Lambda_c) \text{ or } \alpha_{\nu_c} = \alpha_{\min}(\Lambda_c) .$$
 (1.5)

The critical pumping  $\Lambda_c$  is called the second threshold and satisfies  $\Lambda_c \ge \Lambda_{c,\min} \ge 8$ . In the literature this multimode instability is called Risken-Nummedal-Graham-Haken instability. Direct numerical integration of the Maxwell-Bloch laser equations showed that a steady, traveling wave self-pulsing will develop from the RNGH instability, and for certain values of the cavity length the pulsing solution can be stable below the second threshold.<sup>3</sup>

In the vicinity of the second threshold the self-pulsing was analyzed by Haken and Ohno.<sup>5-8</sup> They found analytical results for the temporal form of the laser output and established an analogy between the onset of the self-pulsing and phase transitions in systems far from equilibrium.<sup>9,10</sup>

Later on many authors extended the study to consider systems with detuning<sup>11-13</sup> and systems where the longitudinal profile of the stationary field is not a constant<sup>14</sup> and they found similar multimode instabilities. Other examples can also be found in Refs. 15–18. A comprehensive description of these developments is presented by the review papers;<sup>19-22</sup> see also the introduction of our recent paper.<sup>2</sup> Essentially these kinds of instabilities are caused by the excitation of the Rabi oscillation in laser systems.<sup>22,23</sup> Since the Rabi oscillation is bound to the interaction between the atoms and the electric field, the RNGH instability belongs to an intrinsic property of a multimode laser.

In Ref. 2 we solved analytically the self-pulsing problem in a homogeneously broadened two-level ring laser under the limit  $\gamma_{\parallel} \ll \gamma_{\perp}$ . For this system we found that the onset of the self-pulsing corresponds to a first- (or second-) order phase transition if  $\alpha_{\nu_c} = \alpha_{\max}(\Lambda_c)$  [or  $\alpha_{\nu_c} = \alpha_{\min}(\Lambda_c)$ ]. The physical reason is that a mode near the gain center is suppressed by the stationary laser operation below the second threshold. When this operation becomes unstable and absent, the gain of this mode experiences an additional increase so that the amplitude of the pulsation grows discontinually.

Though the theoretical studies have shown that the RNGH-type instability is intrinsic to a multimode laser, it has not yet been identified experimentally up to now. As pointed out by Abraham, dye lasers are a good candidate for the experimental investigation, since the required cavity mode condition can be easily satisfied.<sup>22</sup>

### B. Multimode instabilities in dye lasers

The experimental observations of the higher-order instabilities and bichromatic operations in dye lasers reported by Hillman *et al.*,<sup>24</sup> and further experiments done by Stroud and co-workers<sup>25-27</sup> have inspired a series of theoretical work to investigate the multimode instabilities in dye lasers.<sup>1,14,28-30</sup>

To explain the novel phenomena observed in the experiments we have proposed a band model for dye lasers<sup>1</sup> in the framework of the semiclassical laser theory developed by Haken.<sup>31,32</sup> The energy-level diagram of this model consists of a single upper lasing level and a lower lasing band of many sublevels. For this model we have found two kinds of multimode instabilities for different population relaxation rates  $\gamma_b$  of the sublevels.

If  $\gamma_b$  is comparable to the relaxation constant  $\gamma_d$  of the upper lasing level, then the occupation of the band cannot be neglected. Through the stimulated emission the sublevels, which are important for a particular laser operation, can be so strongly populated that the corresponding inversions decrease to a certain minimum value. Consequently, the original lasing state becomes unstable and a new laser operation takes place. In contrast to the RNGH instability and self-pulsing this kind of multimode instabilities is caused by saturation of different sublevels and result in new multichromatical laser operations.<sup>29,30</sup> We believe that they are those observed in the experiments.<sup>24–26</sup>

Another case is  $\gamma_b \gg \gamma_d$ , i.e., the population of the sublevels decays very fast and the band is always empty. The stationary solution and its stability in this case have been studied in Ref. 1. The results show that the multimode instability corresponds to a very low second threshold. The physical reason is that the sublevels, which are not in resonance with the stationary laser operation, facilitate the excitation of the side modes.

In this paper we continue to study the case  $\gamma_b \gg \gamma_d$ . We shall show that self-pulsing takes place above the second threshold and present analytical solutions of the pulsations. Since the second threshold and the self-pulsing solutions coincide with those found for two-level systems<sup>2-4</sup> in the limit that the band reduces to a single level, we call this multimode instability RNGH-type instability.

The self-pulsing in the band model has some new distinct features in comparison to that of two-level systems. The most distinct one is that the second threshold may be very low. To observe the RNGH-type instability and the self-pulsing in a dye laser does not require the pump power to be at least nine times the first lasing threshold. For the experimentalist this shows another advantage of using a dye laser to investigate the RNGH-type instability, besides that the cavity mode condition can be easily satisfied.<sup>22</sup> Therefore the self-pulsing solutions presented in this paper may become the first ones to be observed in future experiment on the RNGH instabilities.

Another new feature of the self-pulsing in the band model is that the phase velocity v of the pulsation may be smaller than the light velocity c in the active medium. This means that v > c, which has been regarded as a general property of self-pulsing in active medium,<sup>3</sup> is only an incidental result of the two-level model.

### C. Experimental data of the relaxation constants

The relaxation constants  $\gamma_p$ ,  $\gamma_d$ , and  $\gamma_b$  of the polarization, the population of the upper lasing level, and the population of the band are of particular importance for the band model. For the rhodamine 6G molecule, which is used in the experiments,<sup>24-26</sup>  $\gamma_p$  and  $\gamma_d$  have the typical values  $\gamma_p \simeq 10^{12} - 10^{13} \text{ s}^{-1}$  and  $\gamma_d \simeq 10^9 - 10^{10} \text{ s}^{-1}$ , respectively.<sup>33-35</sup> The measurement of  $\gamma_b$  is very difficult. Ricard and Ducuing measured  $\gamma_b$  for those sublevels which are located spectrally 5600 Å below the upper lasing level and found a value  $\gamma_b \simeq 2.5 \times 10^{11} \text{ s}^{-1}$ .<sup>36</sup> For these sublevels  $\gamma_b \gg \gamma_d$  holds and the population of the sublevels can be neglected.<sup>1</sup> This is the case with which we are concerned in the present work.

We mention in passing that the sublevels which are relevant to the bichromatical operations reported in Ref. 24 are located about 5970 Å spectrally below the upper lasing level. In other words, they lie more than 300 Å lower than the sublevels measured by Ricard and Ducuing. For these lower sublevels the population relaxation constant may be much smaller,<sup>37</sup> as we assumed in Refs. 29 and 30.

The paper is organized as follows. In Sec. II a brief introduction of the band model is presented. The stability of the stationary solution is analyzed in Sec. III. In contrast to Ref. 1, we are interested here in the case of discrete cavity modes because the spacing between adjacent cavity modes in a dye laser is usually comparable to the Rabi frequency. The main part of this work is Sec. IV, where the self-pulsing solutions in the band model are obtained analytically. A summary of the results is given in Sec. V.

### **II. BAND MODEL**

We only present the Maxwell-Bloch equations and explain the meaning of the quantities of the band model. A detailed derivation can be found in Refs. 1 and 23.

The relevant energy-level diagram of a dye laser is described in Fig. 1. It consists of a single upper lasing level, which corresponds to the lowest vibrational level of the



FIG. 1. Relevant energy-level diagram for a cw dye laser consists of an excited single-level and a ground band with many sublevels. The upper lasing level can interact with each of the lower sublevels, but there is not interaction among the sublevels themselves.

first electronic excited state  $(S_1)$  of the dye molecules, and a lower lasing band of many sublevels, which represent the vibrational and rotational levels of the electronic ground state  $(S_0)$  of the molecules. The relevance of these sublevels is evidenced by the very wide spectral tunability (several hundred angstroms) of dye lasers.

Let W and  $|\phi\rangle$  be the eigenvalue and eigenfunction of the excited single level and  $W_m$  and  $|\phi_m\rangle$  be those of the *m*th sublevel, respectively. Then the wave function  $|\Phi\rangle$ of the dye molecule can be expanded as

$$|\Phi\rangle = c(t)e^{-iWt/\hbar}|\phi\rangle + \sum_{m\in\mathcal{B}} c_m(t)e^{-iW_mt/\hbar}|\phi_m\rangle , \quad (2.1)$$

where  $\mathcal{B}$  stands for all the sublevels of the band. Introducing the polarization  $P_m$  with respect to the *m*th sublevel and the population D for the upper level by the correspondence relations,

$$P_{m} \leftrightarrow c(t) c_{m}^{*}(t), \quad D \leftrightarrow |c(t)|^{2}, \qquad (2.2)$$

we can derive the following Maxwell-Bloch equations based on Haken's semiclassical laser theory:<sup>31,32</sup>

$$\frac{\partial E}{\partial t} = -\kappa E - c \frac{\partial E}{\partial z} + \kappa \sum_{m \in \mathcal{B}} P_m , \qquad (2.3)$$

$$\frac{\partial P_m}{\partial t} = -\gamma_p (1 - i\Delta_m) P_m + \gamma_p f_m DE , \qquad (2.4)$$

$$\frac{\partial D}{\partial t} = -\gamma_d \left[ \Lambda + 1 - D - \frac{\Lambda}{2} \sum_{m \in \mathcal{B}} (P_m^* E + P_m E^*) \right] . \quad (2.5)$$

In these equations the population of the sublevels has been neglected due to  $\gamma_b >> \gamma_d$ .<sup>1</sup> E(z,t) refers to the electric-field strength,  $P_m(z,t)$  to the macroscopic polarization with respect to the *m*th sublevel, and D(z,t) to the macroscopic population density of the excited state. They satisfy the periodic boundary conditions

$$E(z+L,t) = E(z,t) ,$$
  

$$P(z+L,t) = P(z,t) ,$$
  

$$D(z+L,t) = D(z,t) .$$
(2.6)

The effective pump parameter  $\Lambda$  in (2.5) is defined by  $\Lambda = (D_0 - D_c)/D_c$ , where  $D_0$  is the unsaturated inversion due to pump and relaxation processes, and  $D_c$  is the threshold inversion to be given in (2.14). By definition the first threshold is  $\Lambda = 0$ .  $\kappa$ ,  $\gamma_p$ , and  $\gamma_d$  are the relaxation rates of E,  $P_m$ , and D, respectively.

The parameters  $\{\Delta_m\}$  are the dimensionless frequency spacing between the *m*th sublevel and the band center m=0.

$$\Delta_m \equiv \frac{W_m - W_0}{\hbar \gamma_p} \quad . \tag{2.7}$$

For simplicity we assume

$$\Delta_m = \Delta m \quad , \tag{2.8}$$

i.e., the sublevels are equally spaced.

The parameters  $\{f_m\}$  characterize the dipole matrix element  $\Theta_m \equiv \langle \phi_m | (-er) | \phi \rangle$  of the  $| \phi \rangle - | \phi_m \rangle$  transition. Define the total dipole moment strength  $\theta$  by

$$\theta \equiv \left[ \sum_{m \in \mathcal{B}} \frac{|\Theta_m|^2}{1 + \Delta_m^2} \right]^{1/2}; \qquad (2.9)$$

then  $f_m$  is defined by

$$f_m \equiv \frac{|\Theta_m|^2}{\theta^2} . \tag{2.10}$$

By definition  $\{f_m\}$  are positive constants and satisfy the normalization

$$\sum_{m \in \mathcal{B}} \frac{f_m}{1 + \Delta_m^2} = 1 .$$
(2.11)

For simplicity, we assume that  $\{f_m\}$  are symmetric with respect to the band center

$$f_m = f_{-m}$$
, (2.12)

and that the central sublevels have greater dipole moments

$$f_0 \ge f_{\pm 1} \ge f_{\pm 2} \ge \cdots > 0$$
. (2.13)

The property (2.13) is a sufficient condition of the stable stationary solution for small pump parameter.<sup>1</sup>

For simplicity the dynamic variables E(z,t),  $\{P_m(z,t)\}$ , and D(z,t) in (2.3)-(2.5) have been normalized by the constants

$$E_{c} = \frac{\hbar\sqrt{\gamma_{d}\gamma_{p}}}{\theta\sqrt{2\Lambda}}, \quad P_{c} = \frac{\hbar\kappa\sqrt{\gamma_{d}\gamma_{p}}}{i\pi\theta\omega_{0}\sqrt{8\Lambda}}, \quad D_{c} = \frac{\hbar\kappa\gamma_{p}}{2\pi\theta^{2}\omega_{0}},$$
(2.14)

where  $\omega_0$  is the frequency of the  $|\phi\rangle - |\phi_0\rangle$  transition.

The stationary solutions of this model and the stability analysis for continuous cavity mode  $(L \rightarrow \infty)$  have been

studied in Ref. 1. In this work we are interested in the case of discrete cavity modes and the self-pulsing solutions developing from the instabilities. It is easy to see that this band model reduces to the two-level model if  $f_0=1$  and  $f_{m\neq 0}=0$ .

## **III. INSTABILITY OF THE STATIONARY SOLUTION**

The stationary solution of (2.3)-(2.5) is given by

$$E_s = 1, \quad P_{m,s} = \frac{f_m}{1 - i\Delta m}, \quad D_s = 1$$
 (3.1)

This solution describes a unidirectional laser operation with the frequency  $\omega_0$ . It is worth pointing out that in this operation not only the center sublevel, which resonates with the lasing field, but all the other sublevels take part in the lasing transitions also.

#### A. General formalism of linear stability analysis

To study the stability we consider small perturbations  $\delta E$ ,  $\delta E^*$ ,  $\delta P_m$ ,  $\delta P_m^*$ , and  $\delta D$  for the stationary solution and make the ansatz

$$(\delta E, \delta E^*, \delta P_m, \delta P_m^*, \delta D) = (e, \tilde{e}, p_m, \tilde{p}_m, d) \exp[\sqrt{\gamma_d \gamma_p} (\lambda t - i\beta z/c)]. \quad (3.2)$$

We call  $\lambda$  and  $\beta$  the eigenvalue and the wave number of the perturbation regardless of the scaling factors. To satisfy the periodic boundary condition  $\beta$  must be equal to 1 of the discrete values

$$\beta_{\nu} = \frac{2\pi c\nu}{L\sqrt{\gamma_d \gamma_p}} \quad (\nu = 0, \pm 1, \pm 2, \ldots)$$
(3.3)

Defining

$$e_{\pm} \equiv e \pm \tilde{e}, \quad p_{m,\pm} = p_m \pm \tilde{p}_m , \qquad (3.4)$$

it is easy to work out two independent characteristic equations for  $(e_{-}, p_{m,-})$  and  $(e_{+}, p_{m,+}, d)$ , respectively. In terms of the band structure function F(x),

$$F(x) \equiv \sum_{m \in \mathcal{B}} \frac{f_m}{1 + i\Delta m + x} , \qquad (3.5)$$

where x is a complex variable, and

$$\gamma \equiv \frac{\gamma_d}{\gamma_p}, \quad \chi \equiv \frac{\kappa}{\gamma_p}$$
 (3.6)

the two independent characteristic equations are given by

$$\frac{\sqrt{\gamma}}{\chi}(\lambda - i\beta) = -1 + F(\sqrt{\gamma}\lambda)$$
(3.7)

and

$$\frac{\sqrt{\gamma}}{\chi}(\lambda - i\beta) = -1 + \frac{(1 + \lambda/\sqrt{\gamma} - \Lambda)F(\sqrt{\gamma}\lambda)}{1 + \lambda/\sqrt{\gamma} + \Lambda F(\sqrt{\gamma}\lambda)} .$$
(3.8)

We shall discuss these two equations separately.

## **B.** Pump-independent characteristic equation

The characteristic equation (3.7) does not depend on the pump parameter A. In the following we show that all the roots of this equation have a nonpositive real part. To this end we need the inequality

$$\operatorname{Re}F(x) = \sum_{m \in \mathcal{B}} \frac{(1+x_r)f_m}{(1+x_r)^2 + (x_i + \Delta m)^2} \le 1$$
for  $x_r \ge 0$  (3.9)

where the equation is valid only for  $x \equiv x_r + ix_i = 0$  ( $x_r$  and  $x_i$  are real). The proof of the inequality for a continuous band was given in Ref. 1. For discrete sublevels the mathematical problem is much more difficult and the proof is given in the Appendix.

Now we consider the real part of (3.7) and show that  $\operatorname{Re}\lambda \leq 0$ . In fact, if  $\operatorname{Re}\lambda > 0$ , then on the left-hand side of the equation one has  $\sqrt{\gamma}\operatorname{Re}\lambda/\chi > 0$ , but on the right-hand side there exists  $-1 + \operatorname{Re}F(\sqrt{\gamma}\lambda) < 0$ , according to (3.9). This contradiction shows  $\operatorname{Re}\lambda \leq 0$ . Furthermore, that the equation in (3.9) exists only for a = b = 0 implies that  $\operatorname{Re}\lambda=0$  exists only for  $\beta=0$ . Obviously all these eigenvalues do not indicate any instability of the stationary solution.

### C. Pump-dependent characteristic equation

We consider (3.8) for a sufficiently small pump at first. At  $\Lambda = 0$  (3.8) reduces to (3.7) if the denominator on the right-hand side is not equal to zero. That is, besides the root  $\lambda = -\sqrt{\gamma}$ , which is the zero of the denominator, all the other roots of (3.8) are equal to those of (3.7). Therefore all the roots for  $\beta \neq 0$  have negative real part and that for  $\beta = 0$  has a zero real part. In the limit  $\Lambda \rightarrow +0$  only the latter may obtain a positive real part. Using perturbation theory one obtains this root (which is zero for  $\beta = \Lambda = 0$ ) from (3.8)

$$\lambda = -\frac{\chi}{\sqrt{\gamma}} \Lambda[2 - F'(0)] < 0 \text{ for } \beta = 0, \Lambda \to +0 \qquad (3.10)$$

where the inequality F'(0) < 0 has been taken into account. The proof of F'(0) < 0 is given in the Appendix. Therefore all the roots of (3.8) have a negative real part (regardless of  $\beta$ ) in the limit  $\Lambda \rightarrow +0$ .

With increasing  $\Lambda$  the stationary solution loses its stability if  $\lambda = i\eta$  ( $\eta$  is real). At this critical point the real part of (3.8) yields

$$\operatorname{Re}\left[\frac{(1+i\eta/\sqrt{\gamma}-\Lambda)F(i\sqrt{\gamma}\eta)}{1+i\eta/\sqrt{\gamma}+\Lambda F(i\sqrt{\gamma}\eta)}\right]=0.$$
(3.11)

Using the auxiliary function

$$H(\eta, \Lambda) \equiv -2\Lambda^2 |F|^2 + \Lambda \left[ |F|^2 - 3F_r - \frac{3\eta F_r}{\sqrt{\gamma}} \right] + \left[ 1 + \frac{\eta^2}{\gamma} \right] (F_r - 1) , \qquad (3.12)$$

where  $F_r$ ,  $F_i$ , and |F| are the real, imaginary part, and ab-

$$H(\eta, \Lambda) = 0 . \tag{3.13}$$

The imaginary part of (3.8) becomes

$$\beta = \eta - \frac{\chi}{\sqrt{\gamma}} \operatorname{Im} \left[ \frac{(1 + i\eta/\sqrt{\gamma} - \Lambda)F(i\sqrt{\gamma}\eta)}{1 + i\eta/\sqrt{\gamma} + \Lambda F(i\sqrt{\gamma}\eta)} \right].$$
(3.14)

The last couple of equations describe the second threshold of the stationary solution. For a two-level atom the band-structure function is given by F(x)=1/(1+x) and (3.13) and (3.14) produce the identical results presented in Sec. I A. Therefore the instability concerned here is an extension of the RNGH instability. We call this instability RNGH-type instability.

Now we solve the threshold condition in the limit

$$\gamma \equiv \frac{\gamma_d}{\gamma_p} \to 0 \tag{3.15}$$

$$\eta_{\pm}(\Lambda) = \left[\frac{1}{|F''|} \{3|F'|\Lambda \pm [9|F'|^2\Lambda^2 - 4|F''|\Lambda(\Lambda+1)]^{1/2}\}\right]$$

These solutions are real only for

$$\Lambda \ge \Lambda_{c,\min} \equiv \frac{1}{9|F'|^2/4|F''|-1} .$$
(3.20)

Therefore  $\Lambda_{c,\min}$  is the minimum value of the second threshold. For a two-level atom one has F(x)=1/(1+x), |F'|=1, and |F''|=2. This produces  $\Lambda_{c,\min}=8$ , a result that one also expects from (1.4) in the limit  $\gamma \rightarrow 0$ . This minimum value was studied in Ref. 1. The numerical results presented there show that  $\Lambda_{c,\min}$  is always smaller than its counterpart of the two-level model and may be equal to any number greater than zero.

In terms of  $\Lambda_{c,\min}$  the critical frequency can be written as

$$\eta_{\pm}(\Lambda) = \left\{ \frac{3|F'|\Lambda}{|F''|} \pm 2 \left[ \frac{\Lambda}{|F''|} \left[ \frac{\Lambda}{\Lambda_{c,\min}} - 1 \right] \right]^{1/2} \right\}^{1/2}.$$
(3.21)

In the same approximation one obtains from (3.14)

$$\beta_{\pm}(\Lambda) = (1 + \chi |F'|) \eta_{\pm}(\Lambda) - \frac{2\chi\Lambda}{\eta_{\pm}(\Lambda)} . \qquad (3.22)$$

In this notation  $\beta_+(\Lambda)$  and  $\beta_-(\Lambda)$  correspond to  $\alpha_{\max}(\Lambda)$ and  $\alpha_{\min}(\Lambda)$ , respectively (see Sec. I A). It is easy to show that

$$\eta_{-}(\Lambda) \leq \eta_{+}(\Lambda), \quad \beta_{-}(\Lambda) \leq \beta_{+}(\Lambda)$$
 (3.23)

where the equalities hold only for  $\Lambda = \Lambda_{c,\min}$ .

The functions  $\beta_{\pm}(\Lambda)$  are the boundaries of the instability region for the stationary solution in the  $(\Lambda,\beta)$  plane. and  $\sqrt{\gamma_d \gamma_p}$  is finite. This is a good approximation for dye lasers, where  $\gamma \simeq 10^{-3}$  and  $\sqrt{\gamma_d \gamma_p} \simeq 10^{11} \text{ s}^{-1}$ ; see Sec. I.C.

In this limit we expand the band structure function into a power series

$$F(i\sqrt{\gamma}\eta) = 1 + F'(0)i\sqrt{\gamma}\eta - F''(0)\frac{\gamma\eta^2}{2} + \cdots \qquad (3.16)$$

Since F'(0) < 0 and F''(0) > 0, where the latter is implied by (3.9), we shall use the abbreviations

$$|F'| = -F'(0), |F''| = F''(0).$$
 (3.17)

Substituting (3.16) in *H*, we obtain

$$H(\eta, \Lambda) = -2\Lambda^{2} + \Lambda(3|F'|\eta^{2} - 2) - \frac{|F''|\eta^{4}}{2} + O(\sqrt{\gamma}) .$$
(3.18)

Neglecting the small term  $O(\sqrt{\gamma})$  we obtain two solutions from (3.13)

For a laser cavity with finite cavity length L only the discrete values  $\{\beta_{\nu}\}$  are allowed; see (3.3). The second threshold  $\Lambda_c$  is the minimum of all possible values fulfilling either of  $\beta_{\nu} = \beta_{\pm}(\Lambda)$ . Denoting the critical integer  $\nu$  by  $\nu_c$  and the critical wave number  $\beta_{\nu_c}$  by  $\beta_c$ , then either

$$\beta_c = \beta_+(\Lambda_c) \text{ or } \beta_c = \beta_-(\Lambda_c)$$
 (3.24)

holds at the second threshold; see Fig. 2. In Sec. IV D we shall show that in the former case the self-pulsing is supercritical, and in the latter case it is subcritical.

### D. Lorentzian distribution of the dipole moments

In order to calculate numerical values of the solutions, we consider a concrete band structure. We assume that the band is continuous and that the distribution of the dipole moments  $\{f_m\}$  is a truncated Lorentzian:

c

$$f(\sigma) = \begin{cases} \frac{C_N}{1 + \sigma^2 / \Gamma^2} & \text{if } |\sigma| \le B_w \\ 0 & \text{if } |\sigma| > B_w \end{cases}$$
(3.25)

where  $\sigma$  corresponds to  $\Delta m$ ,  $2B_w$  is the dimensionless bandwidth (the real bandwidth is equal to  $2B_w \gamma_p \hbar$ ),  $\Gamma$  the halfwidth of the Lorentzian, and  $C_N$  the normalization constant. The density of the sublevels has been included in  $f(\sigma)$ . We can change  $B_w$  and  $\Gamma$  to describe different band structures. For dye molecules the sublevels are very dense and they can be treated as a continuum.<sup>33</sup>

In accord with (2.12) and (2.13)  $f(\sigma)$  satisfies

1



FIG. 2. Unstable region of the stationary solution. At the second threshold one has either (a)  $\beta_c = \beta_+(\Lambda_c)$  or (b)  $\beta_c = \beta_-(\Lambda_c)$ .

$$f(\sigma) = f(-\sigma), \quad f(\sigma) > f(\sigma') \quad \text{if } |\sigma| < |\sigma'| \quad (3.26)$$

The band structure function is given by

$$F(x) = \int_{-B_w}^{B_w} \frac{C_N d\sigma}{(1+\sigma^2)(1+\sigma^2/\Gamma^2)}$$
$$= \frac{C_N}{\Gamma^2 - (1+x)^2} \left[ i\Gamma^2 \ln \frac{1+x-iB_w}{1+x+iB_w} - \frac{2(1+x)}{\Gamma} \arctan \frac{B_w}{\Gamma} \right], \quad (3.27)$$

where the complex logarithmic function should be understood as

$$\ln \frac{1+x-iB_{w}}{1+x+iB_{w}} = \ln \left| \frac{1+x-iB_{w}}{1+x+iB_{w}} \right|$$
$$-i \left[ \arctan \left[ \frac{B_{w}+x_{i}}{1+x_{r}} \right] + \arctan \left[ \frac{B_{w}-x_{i}}{1+x_{r}} \right] \right]. \quad (3.28)$$

The constant  $C_N$  is determined by the normalization condition F(0) = 1 and from (3.27) one obtains

$$C_N = \frac{\Gamma^2 - 1}{2\Gamma^2 [\arctan B_w - (1/\Gamma) \arctan (B_w/\Gamma)]} . \quad (3.29)$$

Straightforward calculations yield

$$F'(0) = \frac{2}{\Gamma^2 - 1} \left[ 1 - \frac{B_w \Gamma^2 C_N}{1 + B_w^2} - \Gamma C_N \arctan \frac{B_w}{\Gamma} \right], \quad (3.30)$$

$$F''(0) = \frac{2}{\Gamma^2 - 1} \left[ 2F'(0) + 1 + \frac{2B_w \Gamma^2 C_N}{(1 + B_w^2)^2} \right].$$
(3.31)

Based on these expressions we have calculated the functions  $\beta_{\pm}(\Lambda)$  and the results are demonstrated in Fig. 2.

Now we point out that this instability requires a cavity length which is usual for dye lasers.<sup>22</sup> To this end we let the critical wave number  $\beta_c = 2$  (compare to Fig. 2) and assume other parameters to be  $c = 3 \times 10^{10}$  cm s<sup>-1</sup>,  $\gamma_d = 10^9$  s<sup>-1</sup>, and  $\gamma_p = 10^{12}$  s<sup>-1</sup>, respectively. It follows then from (3.3) that  $L = 2\pi c / (\beta_c \sqrt{\gamma_d \gamma_p} \approx 3 \text{ cm}.$ 

# IV. SELF-PULSING SOLUTIONS FOR $\gamma \rightarrow 0$

Now we derive the running wave self-pulsing solutions for the band model in the limit (3.15). Since the frequency  $\eta$  of the critical perturbation

$$\exp\left[i\sqrt{\gamma_d\gamma_p}\left[\eta t - \frac{\beta}{c}z\right]\right]$$
(4.1)

lies on the order of 1 [see (3.21)], we introduce the local time variable for the running wave by

$$\tau \equiv \sqrt{\gamma_d \gamma_p} \left[ t - \frac{z}{v} \right] , \qquad (4.2)$$

where v is the phase velocity of the pulsation. By definition we have

$$\frac{\partial}{\partial t} = \sqrt{\gamma_d \gamma_p} \frac{d}{d\tau}, \quad \frac{\partial}{\partial z} = -\frac{\sqrt{\gamma_d \gamma_p}}{v} \frac{d}{d\tau} \quad . \tag{4.3}$$

The scaling of time leads to

$$O\left[\frac{d}{d\tau}\right] = 1 \tag{4.4}$$

for the derivatives of field variables.

For a running wave self-pulsing the Maxwell-Bloch equations (2.3)-(2.5) become

$$\sqrt{\gamma} \epsilon \frac{dE}{d\tau} = -E + \sum_{m \in \mathcal{B}} P_m , \qquad (4.5)$$

$$\sqrt{\gamma} \frac{dP_m}{d\tau} = -(1 - i\Delta m)P_m + f_m ED , \qquad (4.6)$$

$$\frac{dD}{d\tau} = \sqrt{\gamma} \left[ \Lambda + 1 - D - \frac{\Lambda}{2} \sum_{m \in \mathcal{B}} (EP_m^* + E^*P_m) \right], \quad (4.7)$$

where the parameter  $\epsilon$  is defined by

$$\boldsymbol{\epsilon} \equiv \frac{1}{\chi} \left[ 1 - \frac{c}{v} \right] \,. \tag{4.8}$$

### SELF-PULSING IN A BAND MODEL FOR DYE LASERS

In terms of  $\tau$  the periodic boundary condition (2.6) takes the form

$$\begin{bmatrix} E(\tau+T_v), P_m(\tau+T_v), D(\tau+T_v) \end{bmatrix}$$
  
=  $\begin{bmatrix} E(\tau), P_m(\tau), D(\tau) \end{bmatrix}$ , (4.9)

where the period  $T_v$  is defined by

$$T_{v} \equiv \frac{\sqrt{\gamma_{d}\gamma_{p}}L}{v} \quad . \tag{4.10}$$

# A. General solution

We shall solve E,  $\{P_m\}$ , D, and  $\epsilon$  (or v) for given system parameters. To this end we expand  $(E, P_m, D)$  and  $\epsilon$  with respect to the small parameter  $\sqrt{\gamma}$ ,

$$(E, P_m, D) = (E_0, P_{m,0}, D_0) + (E_1, P_{m,1}, D_1)\sqrt{\gamma} + \cdots,$$
(4.11)

$$\epsilon = \epsilon_0 + \epsilon_1 \sqrt{\gamma} + \cdots \qquad (4.12)$$

In order to find the dominant terms  $E_0$ ,  $P_{m,0}$ ,  $D_0$ , and  $D_1$ ( $D_1$  is important to  $E_0$ ) only  $\epsilon_0$  is concerned. For simplicity we denote  $\epsilon_0$  by  $\epsilon$ .

Substituting (4.11) into (4.5)-(4.7), to zeroth order we obtain

$$0 = P_0 - E_0 , \qquad (4.13)$$

$$0 = -(1 - i\Delta m)P_{m,0} + f_m E_0 D_0 , \qquad (4.14)$$

$$\frac{dD_0}{d\tau} = 0 . \tag{4.15}$$

Taking (2.11) into account one obtains

$$P_{m,0} = \frac{f_m}{1 - i\Delta m} E_0, \quad D_0 = 1$$
 (4.16)

The first-order perturbation of (4.5)-(4.7), which determines the field  $E_0(\tau)$ , is given by

$$\epsilon \frac{dE_0}{d\tau} = P_1 - E_1 , \qquad (4.17)$$

$$\frac{dP_{m,0}}{d\tau} = -(1 - i\Delta m)P_{m,1} + f_m(E_1 + E_0D_1), \quad (4.18)$$

$$\frac{dD_1}{d\tau} = \Lambda (1 - E_0 E_0^*) . \qquad (4.19)$$

At first we want to eliminate  $E_1$  and  $P_{m,1}$  from these equations. To this end we divide (4.18) by  $(1 - i\Delta m)$  and then sum over m. This leads to

$$\frac{d}{d\tau} \sum_{m \in \mathcal{B}} \frac{P_{m,0}}{1 - i\Delta m} = -\sum_{m \in \mathcal{B}} P_{m,1} + \sum_{m \in \mathcal{B}} \frac{f_m}{1 - i\Delta m} (E_1 + E_0 D_1) .$$
(4.20)

Considering  $P_{m,0}$  given by (4.16) and the equation

$$\sum_{m \in \mathcal{B}} \frac{f_m}{(1 - i\Delta m)^2} = -F'(0) = |F'| , \qquad (4.21)$$

we obtain

$$|F'|\frac{dE_0}{d\tau} = -P_1 + E_1 + E_0 D_1 . \qquad (4.22)$$

The combination of (4.17) and this equation yields

$$(\epsilon + |F'|) \frac{dE_0}{d\tau} = E_0 D_1$$
 (4.23)

Equations (4.19) and (4.23) form a closed set of equations for  $E_0$  and  $D_1$ . In terms of the intensity variable

$$I \equiv E_0 E_0^*$$
, (4.24)

the two equations take the form

$$(\epsilon + |F'|)\frac{dI}{d\tau} = 2ID_1 , \qquad (4.25)$$

$$\frac{dD_1}{d\tau} = \Lambda(1-I) . \tag{4.26}$$

These equations are similar to those presented in Ref. 2 (for the two-level model |F'|=1). Therefore we simply present the solution as follows. Suppose that the pulse intensity reaches the minimum  $I_{\min}$  at  $\tau=0$ , then the inversion is given by

$$D_{1}(\tau) = \pm \sqrt{\Lambda(\epsilon + |F'|) [\ln I(\tau) - \ln I_{\min} + I_{\min} - I(\tau)]},$$
(4.27)

where the sign is + if  $I(\tau)$  increases with  $\tau$  and it is - if  $I(\tau)$  decreases; see (4.25). It is easy to show that all the (local) minima of the function  $I(\tau)$  are equal to  $I_{\min}$ ; all the maxima are equal to  $I_{\max}$ , which is determined by  $I_{\min}$ :

$$\ln I_{\max} - I_{\max} = \ln I_{\min} - I_{\min} .$$
 (4.28)

The mean value of  $I(\tau)$  can be found by integrating (4.26), which produces

$$\bar{I} = \frac{1}{T_v} \int_0^{T_v} I(\tau) d\tau = 1 .$$
(4.29)

It follows, then, that

$$0 < I_{\min} \le 1 \le I_{\max}$$
, (4.30)

where the equality holds for the stationary solution, which is a limiting case of the self-pulsing. This relation can also be shown by (4.28).

As suggested by (4.27) a real inversion requires that

$$\epsilon + |F'| > 0 . \tag{4.31}$$

This will be shown in (4.49). Now we substitute  $D_1(\tau)$  in (4.25) and obtain

$$d\tau = \pm \frac{1}{2\sqrt{\tilde{\Lambda}}} \frac{dI}{I\sqrt{\ln I - \ln I_{\min} + I_{\min} - I}}, \qquad (4.32)$$

where  $\tilde{\Lambda}$  is defined by

$$\tilde{\Lambda} \equiv \frac{\Lambda}{\epsilon + |F'|} \quad . \tag{4.33}$$

The integration within the least period  $-T/2 \le \tau \le T/2$ leads to the result

$$\tau = \pm \frac{1}{2\sqrt{\tilde{\Lambda}}} \int_{I(\tau)}^{I_{\max}} \frac{dy}{y\sqrt{\ln y - \ln I_{\max} + I_{\max} - y}} .$$
(4.34)

In this period  $I(\tau)$  evolves from  $I_{\text{max}}$  (at  $\tau = -T/2$ ) to  $I_{\text{min}}$  (at  $\tau = I_{\text{min}}$ ) and then back to  $I_{\text{max}}$  (at  $\tau = T/2$ ). Therefore T is given by

$$T = \frac{1}{\sqrt{\tilde{\Lambda}}} \int_{I_{\min}}^{I_{\max}} \frac{dy}{y\sqrt{\ln y - \ln I_{\max} + I_{\max} - y}} \quad (4.35)$$

In order to satisfy the periodic boundary condition (4.9), T must be equal to

$$T_N \equiv \frac{T_v}{N} = \frac{\sqrt{\gamma_d \gamma_p L}}{Nv} , \qquad (4.36)$$

where N is a positive integer. For the right-hand side of (4.35) we have, by (4.28), that

$$\frac{1}{\sqrt{\tilde{\Lambda}}} \int_{I_{\min}}^{I_{\max}} \frac{dy}{y\sqrt{\ln y - \ln I_{\max} + I_{\max} - y}} = \frac{1}{\sqrt{\tilde{\Lambda}}} \int_{I_{\min}}^{I_{\max}} \frac{dy}{\sqrt{\ln y - \ln I_{\max} + I_{\max} - y}} . \quad (4.37)$$

Therefore (4.35) becomes

$$T_{N} \equiv \frac{\sqrt{\gamma_{d}\gamma_{p}L}}{Nv} = \frac{1}{\sqrt{\tilde{\Lambda}}} \int_{I_{\min}}^{I_{\max}} \frac{dy}{\sqrt{\ln y - \ln I_{\max} + I_{\max} - y}}$$
(4.38)

Since within  $z \in (0, L)$  the pulse intensity has N peaks, we call N the pulse number. For later purposes it is helpful to write this equation in the form

$$\frac{c}{v}\sqrt{2\tilde{\Lambda}} = \beta_N \frac{1}{\sqrt{2}\pi} \int_{I_{\min}}^{I_{\max}} \frac{dy}{\sqrt{\ln y - \ln I_{\max} + I_{\max} - y}} ,$$
(4.39)

where  $\beta_N$  is defined in (3.3).

Up to now we have solved (4.5)-(4.7) under the periodic condition (4.9) in the limit  $\gamma \rightarrow 0$ . If  $\epsilon$  and *n* are known, we can calculate  $I_{\min}$  (or  $I_{\max}$ ) for given system parameters from (4.39) and then find  $I(\tau)$  from (4.34).

## B. Determination of the phase velocity v

For the two-level model we have shown that  $\epsilon$  or v, which is the zeroth-order quantity with respect to  $\sqrt{\gamma}$ , is determined by the periodicity of the higher-order expansions of the self-pulsing solution.<sup>2</sup> In doing so one must refer to numerical calculations. In the two-level model we incidentally succeeded to make a correct analytical ansatz for  $\epsilon$ . However, for the band model it is impossible to construct analytical expressions from the numerical calculations. In this paragraph we shall develop an analytical method to determine  $\epsilon$ .

Equation (4.38) shows that for given  $\Lambda$  the phase veloc-

ity does not depend on  $T_N$  or  $I_{\min}$  individually, but depends on their combination. This implies that, for a *fixed*  $\tilde{\Lambda}$ , there are an infinity of solutions which have different  $T_N$  and  $I_{\min}$  (here we consider  $T_N$  as a continuous parameter). A special solution among them is the one whose  $I_{\min}$  approaches 1:

$$I_{\min} = 1 - \delta, \quad \delta \to +0 \;. \tag{4.40}$$

By the approximation

$$\ln(1+\delta) \simeq \delta - \frac{\delta^2}{2} , \qquad (4.41)$$

it follows from (4.28) that

$$I_{\max} = 1 + \delta . \tag{4.42}$$

Supposing

$$I(\tau) = 1 + \delta u(\tau) , \qquad (4.43)$$

we have

$$\sqrt{\ln I(\tau) - \ln I_{\min} + I_{\min} - I(\tau)} = \frac{\delta}{\sqrt{2}} [1 - u^2(\tau)]^{1/2} .$$
(4.44)

Then it follows from (4.34) that

$$\tau = \pm \frac{1}{2\sqrt{\tilde{\Lambda}}} \lim_{I_{\min} \to 1-0} \int_{I_{\min}}^{I(\tau)} \frac{dy}{y\sqrt{\ln y - \ln I_{\min} + I_{\min} - y}}$$
$$= \pm \frac{1}{\sqrt{2\tilde{\Lambda}}} \int_{-1}^{u(\tau)} \frac{du}{(1-u^2)^{1/2}}$$
(4.45)

$$=\pm\frac{1}{\sqrt{2\Lambda}}\left[\arccos u\left(\tau\right)-\pi\right] \tag{4.46}$$

or

$$u(\tau) = -\cos(\sqrt{2\tilde{\Lambda}}\tau) . \qquad (4.47)$$

That is, for given  $\widetilde{\Lambda}$ , among many solutions there is a special pulsation

$$I(\tau) = 1 - \delta \cos \left[ \sqrt{2 \tilde{\Lambda} \gamma_d \gamma_p} \left[ t - \frac{z}{v} \right] \right].$$
 (4.48)

However, since this is the zeroth-order solution with respect to  $\gamma$ , there is no constraint on the frequency  $\sqrt{2\tilde{\Lambda}}$ .

On the other hand, the infinitesimal term in (4.48), which is a periodic but not a damped function of time, is a critical perturbation for the stationary solution. According to the linear stability analysis, which is performed by considering higher-order solutions of the Maxwell-Bloch equations (with respect to  $\sqrt{\gamma}$ ), the frequency of the critical perturbation must be equal to  $\eta_{\pm}(\Lambda)$  as given by (3.21). Therefore, by the definition of  $\tilde{\Lambda}$ , it holds that

$$\left(\frac{2\Lambda}{|F'|+\epsilon}\right)^{1/2} = \eta_{\pm}(\Lambda) \text{ or } \epsilon = \frac{2\Lambda}{\eta_{\pm}^2(\Lambda)} - |F'| . \quad (4.49)$$

4158

In this way we have found  $\epsilon$  for given  $\Lambda$ . We emphasize that this result does not depend on the minimum value  $I_{\min}$ , though it has been derived in the limit  $I_{\min} \rightarrow 1-0$ .

Corresponding to  $\eta_+(\Lambda)$  we denote the two solutions

by  $\epsilon_{\pm}(\Lambda)$ . It is easy to verify that these solutions reduce to those presented in Ref. 2 if a two-level model is concerned, where one has |F'|=1 and |F''|=2.

Using (3.21) we can write the solutions in the form

$$\epsilon_{\pm}(\Lambda) = \frac{2\Lambda}{3\eta_{\pm}^{2}(\Lambda)} \left\{ 1 - \frac{2}{\Lambda_{c,\min}} \mp 2 \left[ \left( \frac{1}{\Lambda_{c,\min}} + 1 \right) \left( \frac{1}{\Lambda_{c,\min}} - \frac{1}{\Lambda} \right) \right]^{1/2} \right\}.$$
(4.50)

The phase velocity v follows from (4.8) and is given by

$$v_{\pm}(\Lambda) = \frac{c}{1 - \chi \epsilon_{\pm}(\Lambda)} . \tag{4.51}$$

These two solutions are shown in Fig. 3.

It is interesting to notice that both the solutions  $\epsilon_{\pm}(\Lambda)$ in the band model may be smaller than zero, e.g., in the case  $\Lambda_{c,\min} < 2$  and  $\Lambda \simeq \Lambda_{c,\min}$  as shown by (4.50). That is, both the possible phase velocities  $v_{\pm}(\Lambda)$  may be smaller than c. Therefore we conclude that the explanation on the relation v > c given in Ref. 3, according to which the inequality v > c must hold generally for self-pulsing, is not correct. v > c is only a result of the two-level model.

In (4.48) we can also consider the spatial behavior of the solution and compare it with the wave number of the critical perturbations. This produces



FIG. 3. Phase velocity v as a function of  $\Lambda$ . It may be greater or smaller than c, depending on the concrete band structure.

$$\frac{c}{v}\sqrt{2\tilde{\Lambda}} = \beta_{\pm}(\Lambda) \tag{4.52}$$

where  $\beta_{\pm}(\Lambda)$  are given by (3.22). It is easy to verify that (4.49) and (4.52) are identical and generate the same results for  $\epsilon_{\pm}(\Lambda)$  and  $v_{+}(\Lambda)$ .

## C. Determination of the pulse number N

From linear stability analysis with respect to the stationary solution we know that for  $\Lambda > \Lambda_c$  the infinitesimal perturbation characterized by  $\beta_c$  becomes unstable, i.e., the corresponding eigenvalue has a positive real part. After the transient growth this unstable mode will develop into a stabilized pulsation. Since there is no other mode which becomes unstable, the pulsation consists of only one basic component, i.e., the unstable mode, and its higher-order harmonics. This implies that the pulsating solution has the same spatial periodicity as that of the unstable mode. Therefore one has for the self-pulsing solution

$$\beta_N = \beta_c \equiv \beta_{\nu_c} \quad \text{or } N = \nu_c \quad . \tag{4.53}$$

## **D.** Final solution

The combination of (4.39), (4.52), and (4.53) produces

$$\beta_{\pm}(\Lambda) = \beta_c \frac{1}{\sqrt{2}\pi} \int_{I_{\min}}^{I_{\max}} \frac{dy}{\sqrt{\ln y - \ln I_{\min} + I_{\min} - y}} \quad (4.54)$$

Now we show that on the left-hand side only  $\beta_+$  yields the right solution. To this end we define the function

$$S(I_{\max}) \equiv \frac{1}{\sqrt{2}\pi} \int_{I_{\min}}^{I_{\max}} \frac{dy}{\sqrt{\ln y - \ln I_{\min} + I_{\min} - y}}$$
. (4.55)

Numerical calculation shows that  $S(I_{\max})$  is a strictly monotonically increasing function of  $I_{\max}$ . The minimum of the function can be calculated analytically and the result is

$$\lim_{I_{\max} \to 1+0} S(I_{\max}) = 1 .$$
 (4.56)

Now we discuss the two possibilities by which the instability of the stationary solution takes place.

In the case  $\beta_c = \beta_+(\Lambda_c)$  it holds that

$$\boldsymbol{\beta}_{-}(\boldsymbol{\Lambda}_{c}) < \boldsymbol{\beta}_{+}(\boldsymbol{\Lambda}_{c}) = \boldsymbol{\beta}_{c} \quad . \tag{4.57}$$

Therefore (4.54) cannot hold if its left-hand side is  $\beta_{-}(\Lambda)$ .

12

In the case  $\beta_c = \beta_-(\Lambda_c)$  it holds that [see Fig. 2(b)]

$$\frac{d\beta_{-}(\Lambda)}{d\Lambda} < 0 \text{ at } \Lambda = \Lambda_c . \tag{4.58}$$

This implies that (4.54) cannot hold for  $\Lambda > \Lambda_c$  if its lefthand side is  $\beta_-(\Lambda)$ . Therefore the only acceptable solution of (4.54) is that given by  $\beta_+(\Lambda)$ .

In terms of  $\eta_{+}(\Lambda)$  and  $\beta_{+}(\Lambda)$  this solution is given by

$$\tau = \pm \frac{1}{\sqrt{2}\eta_{+}(\Lambda)} \int_{I_{\min}}^{I(\tau)} \frac{dy}{\sqrt{\ln y - \ln I_{\min} + I_{\min} - y}} , \quad (4.59)$$

$$\beta_{+}(\Lambda) = \beta_{c} \frac{1}{\sqrt{2}\pi} \int_{I_{\min}}^{I_{\max}} \frac{dy}{\sqrt{\ln y - \ln I_{\min} + I_{\min} - y}} \quad (4.60)$$

For a given system one can solve the pulse amplitude  $I_{\text{max}}$  from (4.60) and then the intensity  $I(\tau)$  from (4.59). The phase velocity of the pulsation is equal to  $v_{+}(\Lambda)$ . This self-pulsing solution is exact for arbitrary  $\Lambda$  in the limit  $\gamma \rightarrow 0$ .

Now we show that the onset of self-pulsing may be continuous (second-order phase transition) or discontinuous (first-order phase transition), depending on how the instability of the stationary solution occurs. We consider (4.60) for the following two cases.

(i)  $\beta_c = \beta_+(\Lambda_c)$ . At the second threshold one has for the self-pulsing  $S(I_{\max}) = 1$ , which yields the same value  $I_{\max} = 1$  as the stationary solution. Therefore the onset of the self-pulsing is continuous. In this case  $I_{\max}$  exists only for  $\Lambda \ge \Lambda_c$  and it increases with increasing  $\Lambda_c$ .

(ii)  $\beta_c = \beta_-(\Lambda_c)$ . Since  $\beta_+(\Lambda_c) > \beta_-(\Lambda_c) = \beta_c$ , it follows that  $S(I_{\max}) > 1$  at  $\Lambda = \Lambda_c$ . Considering the fact that  $S(I_{\max}) > 1$  at  $\Lambda = \Lambda_c$ . Therefore the onset of the self-pulsing is discontinuous when  $\Lambda$  is increased to  $\Lambda_c$ . On the other hand, if  $\Lambda$  is decreased from a value greater than  $\Lambda_c$  by which the self-pulsing already exists, the pulsation persists until  $\Lambda$  is decreased to  $\Lambda_{c,\min}$ , because  $\beta_+(\Lambda_{c,\min}) > \beta_-(\Lambda_c) = \beta_c$  always holds. Thus both the stationary solution and the self-pulsing solution may operate for  $\Lambda \in (\Lambda_{c,\min}, \Lambda_c)$  and the system is bistable. For  $\Lambda < \Lambda_{c,\min}$  no pulse solution is allowed since  $\eta_+(\Lambda)$  and  $\beta_+(\Lambda)$  are no longer real.

The pulse amplitude  $I_{\text{max}}$  as a function of  $\Lambda$  in the two cases is shown in Fig. 4.

Physically the simple criterion about the onset of the self-pulsing may be understood as follows. In case (i) the critical sidemode lies so far from the gain center and thus absorbs so little energy from the system that it maintains only an infinitesimal oscillation at the second threshold. In case (ii) the critical sidemode lies nearer to the gain center, but it is suppressed by the lasing mode (i.e., the stationary solution) whose frequency is located in the gain center. The breaking of the suppression leads to an abrupt increase of gain for the sidemode so that it develops discontinuously into a finite pulsation.



FIG. 4. Pulse amplitude  $I_{max}$  as a function of  $\Lambda$ . (a) If  $\beta_c = \beta_+(\Lambda_c)$ , the self-pulsing solution is supercritical; (b) if  $\beta_c = \beta_+(\Lambda_c)$ , then it is subcritical and the system is bistable for  $\Lambda \in (\Lambda_{c,\min}, \Lambda_c)$ .

## V. CONCLUSIONS AND DISCUSSIONS

We have solved the self-pulsing problem in the band model for ring dye lasers. The analytical solution is given by (4.59) and (4.60), which applies for arbitrary pump parameters. This pulsation resembles that of two-level systems, but it may be realized by a lower pump. Therefore this work suggests a model for the experimental investigation of the RNGH-type instabilities and the pulsations. It should be noted that the stability of the pulse solution has not been studied. Therefore we can only say that, if the laser after the instability of the stationary solution is a traveling wave, then the solution must be given by (4.59) and (4.60).

We have further studied the criterion about the onset of self-pulsing for the two-level model with finite  $\gamma$ . It is found that if  $\beta_c = \beta_-(\Lambda_c)$ , then the onset is discontinuous; if  $\beta_c = \beta_+(\Lambda_c)$ , then it may be discontinuous for  $\beta_c$ smaller than a certain value and it is always continuous for  $\beta_c$  greater than that value. These results show that the criterion in the case of finite  $\gamma$  does not have the simple form as presented in Sec. IV D, but they do support the physical explanation discussed there.

## SELF-PULSING IN A BAND MODEL FOR DYE LASERS

# ACKNOWLEDGMENTS

This research was supported in part by the Volkswagenwerk Foundation, Hannover.

## APPENDIX: INEQUALITIES FOR THE BAND STRUCTURE FUNCTION

Suppose that  $\{f_m\}$   $(m=0,\pm 1,\ldots,\pm N)$  have the properties

$$f_m = f_{-m}, \ f_0 \ge f_{\pm 1} \ge f_{\pm 2} \ge \cdots \ge f_{\pm N} > 0$$
, (A1)

and F(x) is a complex function defined by

$$F(x) \equiv \sum_{m=-N}^{N} \frac{f_m}{1+x-i\Delta m} .$$
 (A2)

Then we have the following two inequalities:

(i) if  $x \equiv x_r + ix_i$  ( $x_r$  and  $x_i$  are real) satisfies  $x \neq 0$  and  $x_r \geq 0$ , then

$$\operatorname{Re}F(0) = \sum_{m=-N}^{N} \frac{f_m}{1 + (\Delta m)^2}$$
  
> 
$$\operatorname{Re}F(x) = \sum_{m=-N}^{N} \frac{f_m(1+x_r)}{(1+x_r)^2 + (x_i - \Delta m)^2} , \quad (A3)$$

(ii)

$$F'(0) = -\sum_{m=-N}^{N} \frac{f_m [1 - (\Delta m)^2]}{[1 + (\Delta m)^2]^2} < 0 .$$
 (A4)

Proof of (i). For simplicity we introduce

$$t_m(x) \equiv \frac{1+x_r}{(1+x_r)^2+(x_i-\Delta m)^2}, \quad m=0,\pm 1,\pm 2,\ldots$$
 (A5)

and

$$S_n(x) = \sum_{m=-n}^{n} t_m(x), \quad n = 0, 1, 2, \dots$$
 (A6)

Now we prove at first that

$$S_n(0) > S_n(x)$$
 for any  $n \ge 0$ . (A7)

Defining  $S_{-1}(0) \equiv S_{-1}(x) \equiv 0$ , it follows from (A6) that

$$S_n(0) - S_n(x) = S_{n-1}(0) - S_{n-1}(x) + R_n(x)$$
 (A8)

From the definitions of  $t_m$  and  $S_n$  one obtains

$$R_{n}(x) = 2t_{n}(0) - t_{-n}(x) - t_{n}(x)$$
$$= \frac{-g_{1}(x)n^{4} - g_{2}(x)n^{2} + g_{3}(x)}{g_{4}(x)}, \qquad (A9)$$

where  $g_j(x)$  (j = 1, 2, 3, 4) are non-negative or positive functions given by

$$g_1(x) \equiv 2\Delta^4 x_r \ge 0 , \qquad (A10)$$

$$g_2(x) \equiv 2\Delta^2[(1+x_r)x_r^2 + (3+x_r)x_i^2] > 0 , \qquad (A11)$$

$$g_3(x) \equiv 2[(1+x_r)^2 + x_i^2][2+x_r)x_r + x_i^2] > 0 , \qquad (A12)$$

$$g_4(x) \equiv [1 + (\Delta m)^2][(1 + x_r)^2 + (x_i - \Delta m)^2] \\ \times [(1 + x_r)^2 + (x_i + \Delta m)^2] > 0 .$$
 (A13)

In (A9) it is obvious that for given x there exists an integer  $K \ge 0$  such that

$$R_n(x) \begin{cases} >0 & \text{if } n < K \\ \ge 0 & \text{if } n = K \\ <0 & \text{if } n > K \end{cases}$$
 (A14)

Since  $R_0(x) = g_3(x)/g_4(x) > 0$ , it follows from (A8) that

$$S_0(0) - S_0(x) = R_0(x) > 0$$
. (A15)

Now we use (A8) and (A14) for n = 0, 1, ..., K, step by step, and find that

$$S_n(0) - S_n(x) > 0$$
 if  $n \le K$ . (A16)

For n > K we have

ſ

$$S_{n-1}(0) - S_{n-1}(x) > S_n(0) - S_n(x)$$
 if  $n > K$ . (A17)

This leads to

$$S_n(0) - S_n(x) > S_{\infty}(0) - S_{\infty}(x)$$
 if  $n > K$ , (A18)

where  $S_{\infty} \equiv \lim_{n \to \infty} S_n$ . The sum of the infinite series can be found by calculating the real part of the residue of the function

$$G(\xi) \equiv \frac{-1}{(1+x-i\Delta\xi)(e^{2\pi i\xi}-1)}$$
(A19)

at the point

$$\tilde{\xi} = \frac{1+x}{i\Delta} \quad . \tag{A20}$$

That is

$$S_{\infty}(x) = \operatorname{Re} \sum_{m=-\infty}^{\infty} \frac{1}{1+x-i\Delta m} = \operatorname{Re} \oint_{\overline{\xi}} G(\xi) d\xi$$
$$= \left[\frac{a}{2}\right] \frac{\sinh(a+b)}{\cosh(a+b)-\cos c} , \qquad (A21)$$

where a, b, and c are given by

$$a \equiv \frac{2\pi}{\Delta} \neq 0, \quad b \equiv \frac{2\pi x_r}{\Delta}, \quad c \equiv \frac{2\pi x_i}{\Delta}.$$
 (A22)

It then follows

$$S_{\infty}(0) = \left| \frac{a}{2} \right| \frac{\sinh a}{\cosh a - 1}$$
 (A23)

Therefore we have, by simple calculations,

$$S_{\infty}(0) - S_{\infty}(x) = \left(\frac{a}{2}\right) \frac{\sinh(a+b) - \sinh b - \sinh a \cos c}{(\cosh a - 1) [\cosh(a+b) - \cos c]} .$$
(A24)

By the definitions of a and b and the assumption  $x_r \ge 0$ we have  $a \ne 0$  and  $a + b = a(1+x_r) \ne 0$ . Therefore the denominator (A24) is positive because  $\cosh > 1$  for any real and nonzero argument.

Now we show that the numerator of (A24) is also positive. Since the numerator is an even function of  $\Delta$ , we may assume, without loss of generality, that  $\Delta > 0$ , i.e., a > 0 and  $b \ge 0$ . Then we have

 $\sinh(a+b) - \sinh b - \sinh a \cos c$ 

$$\geq \sinh(a+b) - \sinh b - \sinh a$$

= sinha coshb + sinhb cosha - sinha - sinhb  $\geq 0$ .

(A25)

This means

$$S_{\infty}(0) - S_{\infty}(x) \ge 0 . \qquad (A26)$$

It follows then from (A18) that

$$S_n(0) - S_n(x) > 0$$
 if  $n > K$ , (A27)

(A16) and (A27) are identical to (A7).

Using (A7) we can prove (A3). To this end we use the transformation

$$\sum_{m=-N}^{N} f_m t_m = \sum_{n=0}^{N-1} (f_n - f_{n+1}) \sum_{m=-n}^{n} t_m + f_N \sum_{m=-N}^{N} t_m .$$
(A28)

From (A1) and (A7) it follows that

$$\sum_{m=-N}^{N} f_m t_m(0) - \sum_{m=-N}^{N} f_m t_m(x)$$
  
= 
$$\sum_{n=0}^{N-1} (f_n - f_{n+1}) [(S_n(0) - S_n(x)] + f_N [(S_N(0) - S_N(x)] > 0.$$
(A29)

We have thus proved the inequality (A3).

Proof of (ii). The inequality (A4) can be written as

$$\sum_{m=-N}^{N} \frac{2f_m}{[1+(\Delta m)^2]^2} - \sum_{m=-N}^{N} \frac{f_m}{1+(\Delta m)^2} > 0 .$$
 (A30)

To prove it we introduce

$$U_n \equiv \sum_{m=-n}^{n} \frac{2}{\left[1 + (\Delta m)^2\right]^2}, \quad V_n \equiv \sum_{m=-n}^{n} \frac{1}{1 + (\Delta m)^2} .$$
(A31)

Considering (A28) and (A29), we find that (A30) holds if and only if

$$U_n - V_n > 0 \quad \text{for any } n \ge 0 \ . \tag{A32}$$

This inequality can be proved by applying the same approach which has been used to prove (A7). Similar to (A8) we define  $\tilde{R}_n$  by

$$U_n - V_n = U_{n-1} - V_{n-1} + \tilde{R}_n$$
, (A33)

where  $U_{-1} \equiv V_{-1} \equiv 0$ . It is easy to find that

$$\tilde{R}_{n} = 2 \frac{1 - (\Delta n)^{2}}{[1 + (\Delta n)^{2}]^{2}} .$$
(A34)

Obviously  $\widetilde{R}_n$  has the property (A14). Therefore the sufficient condition for (A32) is that

$$U_{\infty} - V_{\infty} > 0 , \qquad (A35)$$

where  $U_{\infty} \equiv \lim_{n \to \infty} U_n$  and  $V_{\infty} \equiv \lim_{n \to \infty} V_n$ . The summations can be performed by calculating the residues of certain functions and the final result is

$$U_{\infty} - V_{\infty} = \frac{a^2}{2\sinh^2 a} > 0, \quad a \equiv \pi/\Delta$$
 (A36)

This proves (A35) and hence the inequality (A4).

<sup>1</sup>Hong Fu and H. Haken, Phys. Rev. A 36, 4802 (1987).

- <sup>2</sup>Hong Fu, Phys. Rev. A **40**, 1868 (1989).
- <sup>3</sup>H. Risken and K. Nummedal, J. Appl. Phys. **39**, 4662 (1968).
- <sup>4</sup>R. Graham and H. Haken, Z. Phys. **213**, 420 (1968).
- <sup>5</sup>H. Haken, Z. Phys. B 21, 105 (1975).
- <sup>6</sup>H. Haken and H. Ohno, Opt. Commun. 16, 205 (1976).
- <sup>7</sup>H. Ohno and H. Haken, Phys. Lett. **59A**, 261 (1976).
- <sup>8</sup>H. Haken and H. Ohno, Opt. Commun. 26, 117 (1978).
- <sup>9</sup>H. Haken, Synergetics—An Introduction, 3rd ed. (Springer-Verlag, Berlin, 1983).
- <sup>10</sup>H. Haken, Advanced Synergetics (Springer-Verlag, Berlin, 1987).
- <sup>11</sup>J. Zorell, Opt. Commun. **38**, 127 (1981).
- <sup>12</sup>L. M. Narducci, J. R. Tredicce, L. A. Lugiato, N. B. Abraham, and D. K. Bandy, Phys. Rev. A 33, 1842 (1986).
- <sup>13</sup>L. A. Lugiato, D. K. Bandy, L. M. Narducci, J. R. Tredicce, H. Sadiky, and N. B. Abraham, in *Optical Bistability III*, edited by H. M. Gibbs, P. Mandel, N. Peyghambarian, and S. D. Smith (Springer, Heidelberg, 1986), p. 293.
- <sup>14</sup>L. A. Lugiato, L. M. Narducci, E. V. Eschenazi, D. K. Bandy, and N. B. Abraham, Phys. Rev. A 32, 1563 (1986).
- <sup>15</sup>J. Opt. Soc. Am. B 2, special issue on instabilities in active op-

tical media (1985).

- <sup>16</sup>Optical Instabilities, Proceedings of the International Meeting on Instabilities and Dynamics of Lasers and Nonlinear Optical Systems, edited by R. W. Boyd, M. G. Raymer, and L. M. Narducci (Cambridge University Press, Cambridge, England, 1986).
- <sup>17</sup>Lasers and Synergetics, Vol. 19 of Springer Proceedings in Physics, edited by R. Graham and A. Wunderlin (Springer-Verlag, Berlin, 1987).
- <sup>18</sup>J. Opt. Soc. Am. B 5, special issue on nonlinear dynamics of lasers (1988).
- <sup>19</sup>R. Harrison and D. J. Biswas, Prog. Quantum Electron. 10, 3 (1985).
- <sup>20</sup>P. W. Milonni, M. L. Shih, and J. R. Ackerhalt, *Chaos in Laser-Matter Interactions* (World Scientific, Singapore, 1987).
- <sup>21</sup>N. B. Abraham, P. Mandel, and L. M. Narducci, Prog. Opt. 25, 1 (1987).
- <sup>22</sup>M. L. Narducci and N. B. Abraham, *Laser Physics and Laser Instabilities* (World Scientific, Singapore, 1988), Sec. IV.G.
- <sup>23</sup>Hong Fu, Ph.D. Universität Stuttgart, 1989.
- <sup>24</sup>L. W. Hillman, J. Krasinski, R. W. Boyd, and C. R. Stroud, Jr., Phys. Rev. Lett. **52**, 1605 (1984).

- <sup>25</sup>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 University Press, Cambridge, England, 1986), p. 274.
- <sup>26</sup>C. R. Stroud, Jr., K. Koch, S. Chakmakjian, and L. W. Hillman, in *Optical Chaos*, edited by J. Chrostowski and N. B. Abraham (SPIE, Bellingham, WA, 1986), Vol. 667, p. 48.
- <sup>27</sup>L. W. Hillman, J. Krasinski, R. W. Boyd, and C. R. Stroud, Jr., J. Opt. Soc. Am. B 2, 211 (1985).
- <sup>28</sup>L. W. Hillman and K. Koch, in *Optical Instabilities* (Ref. 25), p. 256.
- <sup>29</sup>Hong Fu and H. Haken, J. Opt. Soc. Am. B 5, 899 (1988).
- <sup>30</sup>Hong Fu and H. Haken, Phys. Rev. Lett. **60**, 2614 (1988).
- <sup>31</sup>H. Haken, Laser Theory, Vol. XXV/2c of Encyclopedia of Physics (Springer-Verlag, Berlin, 1984).
- <sup>32</sup>H. Haken, Light, Vol. 2, Laser Light Dynamics (North-

Holland, Amsterdam, 1985).

- <sup>33</sup>F. P. Schäfer, in *Dye Lasers*, edited by F. P. Schäfer (Springer-Verlag, Berlin, 1973), Chap. 1.
- <sup>34</sup>H. E. Lessing and A. Von Jena, in *Laser Handbook*, edited by M. L. Stitch (North-Holland, Amsterdam, 1979), Vol. 3, p. 6.
- <sup>35</sup>A. Seilmeier and W. Kaiser, in Ultrashort Laser Pulses and Applications, Vol. 66 of Topics in Applied Physics, edited by W. Kaiser (Springer-Verlag, Berlin, 1988), p. 279.
- <sup>36</sup>D. Ricard and J. Ducuing, J. Chem. Phys. 62, 3616 (1975).
- <sup>37</sup>H. Graener, R. Dohlus and A. Laubereau, in Ultrafast Phenomena V, Vol. 46 of Springer Series in Chemical Physics, edited by G. R. Fleming and A. E. Siegman (Springer-Verlag, Berlin, 1986), p. 458.
- <sup>28</sup>L. W. Hillman and K. Koch, in Optical Instabilities (Ref. 25), p. 256.