Optical bistability in a dye-ring cavity

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The optical bistability in a dye-ring cavity has been investigated by using the derived Maxwell-Bloch equations based on the band model. It is shown that the bistable conditions and behavior are closely related to the band structure of the dye molecules.

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I. INTRODUCTION

A dye laser which is easily tunable over a wide range of frequencies has been of considerable experimental [1,2] and theoretical [3–5] interest. Recently, Fu and Haken proposed a band model of the dye molecules [3]. In this model, an organic dye molecule is assumed to have an effective electron and an energy-level diagram consisting of a continuous bandlike ground state with many vibronic sublevels and an excited singlet state (see Fig. 1). The excited singlet state can interact with the vibronic sublevels of the ground state, but there is no interaction among the vibronic sublevels themselves. This band-model theory gives an acceptable explanation for the recent experiments done by Hillman *et al.* [1].

Although the optical bistable behavior of dye systems has been studied in some papers by using a two-level [6], four-level [7], or six-level model [8], it is necessary to conduct further investigation according to the bandlike feature of the dye molecular-energy structure.

In this paper, we investigate theoretically the optical bistability in a unidirectional cavity (Fig. 2) in terms of the band model. For simplicity, we assume that mirrors 3 and 4 have 100% reflectivity, and call R and T (with $R+T=1$) the reflection and transmission coefficient of mirrors 1 and 2. We describe the dynamics of the coupled system (molecules plus radiation field) by the Maxwell-Bloch equations. We obtain that the bistability can be realized in a wide frequency range.

The paper is organized as follows. The Maxwell-Bloch equations and the boundary conditions for the unidirectional cavity are presented in Sec. II, the general bistability is studied in Sec. III, the bistable behavior for some situations is analyzed in Sec. IV, and a summary and conclusions are given in Sec. V.

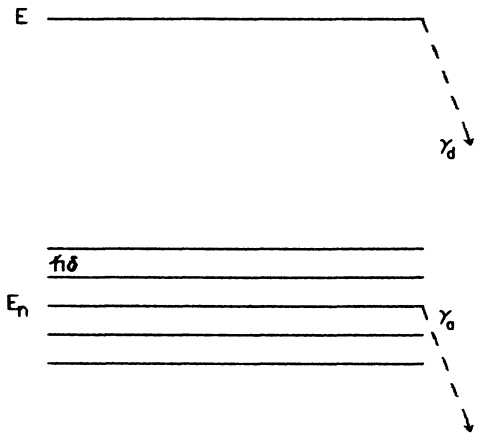

FIG. 1. Relevant energy diagram for an ideal dye molecule. E and E_n represent, respectively, the singlet excited state and sublevels of the ground state, $\hbar\delta$ is the sublevel spacing, γ_d and γ_a denote the decay rates.

II. MAXWELL-BLOCH EQUATIONS

Let E (E_n) and $|\phi\rangle$ ($|\phi_n\rangle$) be the eigenvalue and eigenfunction of the single excited state (nth sublevel of the ground state). The wave function $|\Phi\rangle$ of a dye molecule can be expanded as

$$|\Phi\rangle = c(t)|\phi\rangle + \sum_n c_n(t)|\phi_n\rangle \ . \tag{1}$$

The Schrödinger equation for a molecule with an effective election in an applied electric field $F(z,t)$ reads as

$$i\hbar\dot{c}(t) = Ec(t) - \sum_n c_n(t)\theta_n F(z,t) \ , \tag{2}$$

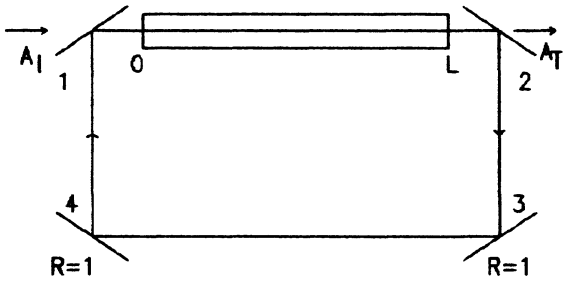

FIG. 2. Ring cavity. A_I and A_T are the incident and transmitted fields, respectively.

$$i\hbar\dot{c}_n(t)=E_n c_n(t)-c(t)\theta_n^* F(z,t) , \tag{3}$$

where $F(z,t)$ is assumed to be quasi monochromatic, i.e.,

$$F(z,t)=A(z,t)\cos(\omega t-kz), \quad k=\omega/c , \tag{4}$$

in which $A(z,t)$ is the slowly varying amplitude of $F(z,t)$, $\theta_n=-\langle\phi|ez|\phi_n\rangle$ is the transition dipole moment from the nth sublevel to the excited state, and θ_n^* is its complex conjugate. We can write [3]

$$\theta_n\theta_n^*=f_n\theta^2 , \tag{5}$$

where the positive number θ stands for the typical amplitude of the dipole matrix element, the distribution function f_n is assumed to be symmetric about the zeroth sublevel (i.e., about the center of the band) [3], namely $f_n=f_{-n}$ ($n=0,\pm1,\ldots,\pm M$ with $2M+1$ being the total number of the sublevels), and is monotonic,

$$f_0\geq f_1\geq\cdots\geq f_m>0 . \tag{6}$$

The electric field $F(z,t)$ satisfies the Maxwell wave equation

$$\nabla^2F-\frac{1}{c^2}\frac{\partial^2F}{\partial t^2}=\frac{4\pi}{c^2}\frac{\partial^2P}{\partial t^2} , \tag{7}$$

where P is the polarization (dipole moment per unit volume) which is determined by the Schrödinger equation. Thus the light-matter-interaction problem can be solved self-consistently by combining the Schrödinger equation [Eqs. (2) and (3)] and the Maxwell equation [Eq. (7)].

In order to facilitate the following study, we define the modified Bloch variables:

$$x_n=c(t)c_n^*(t)\theta_n^*+c^*(t)c_n(t)\theta_n , \tag{8}$$

$$y_n=i[c(t)c_n^*(t)\theta_n^*-c^*(t)c_n(t)\theta_n] , \tag{9}$$

$$z_n=c_n(t)c_n^*(t)\theta_n\theta_n^* , \tag{10}$$

$$w=c(t)c^*(t) . \tag{11}$$

From Eqs. (2)–(4) and (7)–(11), by using the slowly-varying-envelope assumption and by taking into account phenomenologically the molecular relaxation, we obtain the following Maxwell-Bloch equations

$$\dot{u}_n=-\beta u_n+(\Delta_n-\Delta)v_n , \tag{12}$$

$$\dot{v}_n=-\beta v_n-(\Delta_n-\Delta)u_n+\frac{f_n\theta^2A}{\hbar}(w-w_n) , \tag{13}$$

$$\dot{w}_n=-\gamma_a w_n+\frac{A}{2\hbar}v_n , \tag{14}$$

$$\dot{w}=-\gamma_d(1+w)-\frac{A}{2\hbar}\sum_n v_n , \tag{15}$$

$$\dot{A}=-c\frac{\partial A}{\partial z}+2\pi N\omega\sum_n v_n , \tag{16}$$

where $w_n=c_n(t)c_n^*(t)=z_n/f_n\theta^2$, u_n and v_n are the slowly varying amplitudes given by

$$x_n=u_n\cos(\omega t-kz)-v_n\sin(\omega t-kz) , \tag{17}$$

$$y_n=v_n\cos(\omega t-kz)+u_n\sin(\omega t-kz) , \tag{18}$$

N is the number density of dye molecules, $\Delta_n=\omega_0-\omega_n=(E_n-E_0)/\hbar$ is the detuning of the nth sublevel from the central sublevel, $\Delta=\omega_0-\omega$ is the detuning of the field-carrier frequency ω from ω_0, and β, γ_a, and γ_d are, respectively, the relaxation rates for u_n (v_n), w_n, and w. Here we have assumed that the dye molecules are all in their ground state when the field is absent [i.e., we have written $w_0=-1$ in Eq. (15)] and have assumed the loss to occur only at the mirrors (i.e., the cavity decay is negligible). In obtaining Eqs. (12) and (13), we have used the relation $c_n(t)c_m^*(t)$ $(m\neq n)=0$, which has been justified by Ref. [3].

The coherent field A_I enters into the cavity from the left and drives the dye molecules. For a perfectly tuned cavity, the relations between A_I, and the transmitted field A_T, and the fields $A(0)$ and $A(L)$ are [9]

$$A(L)=A_T/\sqrt{T} , \tag{19}$$

$$A(0)=\sqrt{T}\,A_I+R\,A(L) , \tag{20}$$

where L is the length of the molecular sample, and the second contribution on the right-hand side of Eq. (20) describes a feedback mechanisms due to the mirror which is essential to give rise to bistability, namely, there will be no bistability if $R=0$.

III. STEADY-STATE BEHAVIOR

We assume for simplicity that the sublevels have equal spacing $\hbar\delta$, then $\Delta_n=n\delta$. Now we consider here the situation $|\Delta|\leq M\delta$, i.e., the field frequency ω is resonant with the molecular transition frequency from some nth sublevel to the excited singlet state.

Setting all time derivatives in Eqs. (12)–(16) equal to zero for the steady state, after simple calculations, we obtain the normalized field amplitude A' as follows:

$$\frac{dA'}{dz}=-\alpha\frac{A'g}{1+\frac{1}{2}A'^2g} , \quad A'\equiv\frac{\theta}{\hbar(\beta\gamma_d)^{1/2}}A , \tag{21}$$

where $\alpha=2\pi Nk\theta^2/\hbar\beta$ is a typical linear absorption coefficient per unit length, g is the resonant factor defined by

$$g=\sum_n\frac{f_n}{1+(\Delta_n'-\Delta')^2+\frac{1}{2}\gamma f_n A'^2} , \tag{22}$$

which depends on both the field frequency detuning (Δ') and field intensity (A'^2) and is a monotonically decreasing function of A'^2 and Δ' (in proving this fact, we have used the monotonic property of f_n [Eq. (6)]). Here we have introduced the notation

$$\Delta'\equiv\Delta/\beta , \quad \Delta_n'\equiv\Delta_n/\beta, \quad \gamma\equiv\gamma_d/\gamma_a . \tag{23}$$

In the mean-field limit [9] $A'(z)$ is assumed to depend slowly on z so that

$$\int_0^L dz \frac{A'(z)g(\Delta', A'(z))}{1+\frac{1}{2}A'^2(z)g(\Delta', A'(z))} \cong L\frac{A'(L)g(\Delta', A'(L))}{1+\frac{1}{2}A'^2(L)g(\Delta', A'(L))} . \quad (24)$$

Using the boundary conditions (19) and (20), we obtain the mean-field state equation

$$y = x + \frac{2Cxg(\Delta', x)}{1+\frac{1}{2}x^2 g(\Delta', x)} ,$$
$$x \equiv \frac{\theta A_T}{\hbar(\beta\gamma_d T)^{1/2}} , \quad y \equiv \frac{\theta A_I}{\hbar(\beta\gamma_d T)^{1/2}} , \quad C \equiv \frac{\alpha L}{2T} . \quad (25)$$

We can see from Eq. (25) that this optical system can lead to bistability. Generally, the bistable behavior relates to the band structure.

Equation (25) can be rewritten as

$$Y = X[1+Cf(\Delta', X)]^2 , \quad (26)$$

where $Y=y^2$, $X=x^2$, and

$$f(X,\Delta') = \frac{2g(\Delta', X)}{1+\frac{1}{2}Xg(\Delta', X)} . \quad (27)$$

The critical point (at the onset of bistability) is given by $dY/dX = d^2Y/dX^2 = 0$ [10], hence the bistable threshold can be solved as follows:

$$3f'(X_C)+2X_C f''(X_C)=0 , \quad (28)$$

$$C_{\text{crit}} = -[f(X_C)+2X_c f'(X_C)]^{-1} . \quad (29)$$

The optical bistability exists when $C > C_{\text{crit}}$. Equations (28) and (29) can be numerically solved and lead to the results for the critical threshold as a function of Δ' and of the band-structure parameters (f_n, Δ_n). In the next section, we will discuss a typical situation of the system.

IV. THE BISTABLE BEHAVIOR FOR $\gamma \ll 1$

Now we consider the situation for $\gamma \ll 1$, which means that the population of the sublevels decays much faster than that of the excited singlet state [3,5]. In this case w_n are much smaller than w, u_n, v_n, and A in Eqs. (12)–(16) and can be ignored. Equations (25) and (22) then become

$$y = x + \frac{2Cxg(\Delta')}{1+\frac{1}{2}x^2 g(\Delta')} , \quad (30)$$

$$g(\Delta') = \sum_n \frac{f_n}{1+(\Delta'_n-\Delta')^2} . \quad (31)$$

It is shown that $g(\Delta')=1$ as $\Delta'=0$, and $g(\Delta')<1$ as $\Delta'\neq 0$ [3].

Using Eqs. (28) and (29), we obtain

$$C_{\text{crit}} g(\Delta') = 4 . \quad (32)$$

This is the bistable threshold condition for the dye-ring cavity. From Eqs. (31) and (32), we can see that bistability is impossible for $C<4$; for a fixed value of $C>4$, bistability exists in a finite domain of Δ'. Equation (31) shows that when Δ' increases, $g(\Delta')$ decreases much more slowly than its counterpart in a two-level system (i.e., $1/(1+\Delta'^2)$), hence for a given C (>4), Eq. (32) can be satisfied in a much larger domain of Δ'. In other words, bistability in a dye-ring system can be realized in a much wider frequency range.

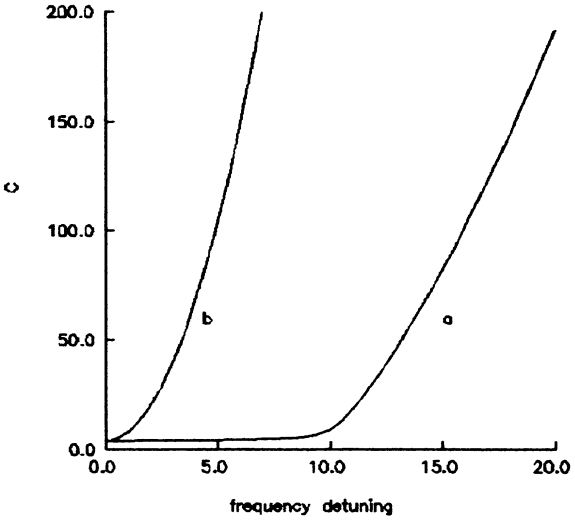

FIG. 3. The plot of C ($\equiv \alpha L/2T$) vs Δ' (frequency detuning) for the band parameters $\Gamma=20.0$ and $\xi=10.0$. This set of parameters is used for Figs. 3 and 4. Curve a corresponds to the band model, and curve b to the two-level model.

In the following numerical analysis, we assume that the band is continuous, and that the distribution of dipole moments f_σ, as a function of σ, be a Lorentzian function of width Γ, i.e.,

$$f(\sigma) = \frac{\text{const}}{1+\sigma^2/\Gamma^2} , \quad (33)$$

where const is to be determined by the normalization condition $g(0)=1$. Equation (32) then becomes

$$C\int_{-\xi}^{\xi} \frac{\text{const}}{[1+(\sigma-\Delta')^2](1+\sigma^2/\Gamma^2)} d\sigma = 4 , \quad (34)$$

where ξ is the half-width of the band.

Setting the band parameter $\Gamma=20.0$, $\xi=10.0$, we obtain the plot of C versus Δ' as shown in Fig. 3. In this figure, curve a corresponds to Eq. (34) (for the band model), whereas curve b is the counterpart in the two-level model [corresponding to $f(\sigma)=\delta(\sigma)$ in Eq. (33)]. These two curves coincide at the point $\Delta'=0$, $C=4$. Since for a given C (>4), bistability can be realized when $|\Delta'|<\Delta_c'$, we can see clearly from the figure that the frequency range for the realization of bistability in the band model is much wider than that of the two-level model. For example, if we set $C=10.0$, the bistability can occur within the whole half-width ($|\Delta'|\leq 10.0$) in the band model, but in the two-level model, $|\Delta'|\leq 1.2$.

Setting the same band parameters as for Fig. 3, we obtain the plot of transmitted light versus incident light as shown in Fig. 4, in which curve a represents the bistable curve based on Eq. (30) for $C=150.0$, $\Delta'=5.0$; curve b is

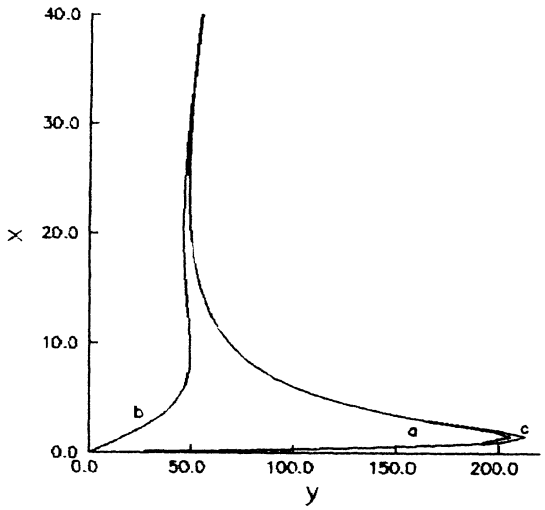

FIG. 4. The plot of transmitted light vs incident light; (curve *a*) for the band model with $C=150.0$ and $\Delta'=5.0$, and (curve *b*) for the two-level model with the same C and Δ'. Curve *c* is the bistable curve for both models with $C=150.0$ $\Delta'=0.0$.

the counterpart for the two-level model with the same C and Δ'; and curve *c* is the bistable curve for both models with $\Delta'=0.0$, $C=150.0$. Figures 3 and 4 clearly show that when $\Delta'=0$, the bistable threshold condition and the bistable curve in the band model are identical with that of the two-level system; but when $\Delta'\neq 0$, in the two-level system the bistable threshold value C_{crit} strongly depends on Δ' and the range of transmitted light in the hysteresis cycle is very narrow (curve *b* in Fig. 3), whereas in the case of the band model, within the limit $|\Delta'|\leq 10.0$, the threshold value C_{crit} is nearly a constant (curve *a* in Fig. 3), and the bistable hysteresis curve is very close to the largest cycle (corresponding to $\Delta'=0$) as shown by curve *c* in Fig. 4.

V. CONCLUSION

In this paper, we have derived the Maxwell-Bloch equations for a dye optical system based on the band model. By using these equations, we have described the optical bistability in the framework of the mean-field limit, and have shown that, in the case of Δ' smaller than the half-width of the band, the bistable threshold value C_{crit} is nearly a constant and the hysteresis is very close to the largest cycle, so we can see clearly that the bistability can be realized in a wide frequency range.

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[1] L. H. Hillman, J. Krasinki, R. W. Royd, and C. R. Stroud, Jr., Phys. Rev. Lett. **52**, 1605 (1984).
[2] N. M. Lawandy, R. Sohrab Afzal, and W. S. Rabinovich, Phys. Rev. A **36**, 1759 (1987).
[3] Hong Fu and H. Haken, Phys. Rev. A **36**, 4802 (1987).
[4] Hong Fu and H. Haken, Phys. Rev. Lett. **60**, 2614 (1988).
[5] Hong Fu and H. Haken, Phys. Rev. A **42**, 4151 (1990).
[6] L. Mandel, Rajarshi Roy, and Surendra Singh, in *Optical Bistability,* edited by Charles M. Bowden, Mikael Ciftan, and Hermann R. Robl (Plenum, New York, 1981), p. 127.
[7] Zhen Fu Zhu and E. Garmire, in *Optical Bistability 2,* edited by Charles M. Bowden, Hyatt M. Gibbs, and Samuel L. McCall (Plenum, New York, 1984), p. 337.
[8] A. Baczynski, A. Kossakowski, and T. Marszalek, Z. Phys. B **23**, 205 (1976).
[9] R. Bonifacio and L. A. Lugiato, Lett. Nuovo Cimento **21**, 505 (1978).
[10] P. D. Drummond, IEEE J. Quantum Electron. QE-**17**, 301 (1981).