Dynamics of homogeneously broadened lasers: higher-order bichromatic states of operation

Lloyd W. Hillman,* Jerzy Krasinski,† Karl Koch, and C. R. Stroud, Jr.

Institute of Optics, University of Rochester, Rochester, New York 14627

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A theory is presented that describes a series of bichromatic states of an ideal homogeneously broadened laser. These states are produced when the laser is pumped far above threshold so that the intracavity field is strong enough to overcome damping and produce Rabi oscillations of the atomic inversion. The two-frequency laser field is amplitude modulated at a frequency that may be either the Rabi frequency or a subharmonic of it. The theoretical predictions are compared qualitatively with recent experimental results.

1. INTRODUCTION

The coupled nonlinear equations that describe the operation of a laser, which include the full coherent atomic dynamics and the many modes of the optical field, are far too complicated to solve generally.[1] A common approach to such complex problems is to assume some particular type of solution, to make appropriate approximations, and to find self-consistent solutions to these approximate equations. One can then carry out a linear stability analysis of the self-consistent solutions to see whether the system will operate stably in the given state. Generally, the stability analysis shows that the system will operate stably only within some limited range of the parameters. Outside this range, these solutions are unstable. In a real system, there are inevitable thermodynamic fluctuations that may further limit the range of stability. Beyond the range of the stable solution, there may or may not exist other stable solutions. A linear stability analysis can never determine whether there are new stable solutions of a type different from those initially assumed.

A linear stability analysis of an ideal homogeneously broadened laser operating in a single mode shows that when the laser is pumped far above threshold, it can become unstable to the growth of new spectral components of the field symmetrically displaced about the initial frequency. This instability was noted some 20 years ago.[2,3] What was not predicted was the nature of the new stable states, if any, beyond the threshold of this instability. A number of quite plausible possibilities exist: (1) A stable three-frequency amplitude or phase-modulated state might exist, (2) further sidebands might develop, leading to self-mode-locked operation, or (3) the two sidebands might simply quench the single mode and produce stable two-frequency operation. The linear stability analysis of the initial single-mode state cannot help us to choose from among these possibilities. On the other side of the stability limit, the neglected nonlinear terms contribute to the damping and saturation that are necessary in establishing a new stable state.

One approach to this problem is simply to put rather general equations into a computer and study the time evolution of the system. Indeed, this has been done by a series of investigators studying spontaneous mode locking.[4] This method has shown the existence of instabilities and predicted the appearance of very-short-pulse solutions for lasers operating far above threshold. However, the parameter space is simply too large for a comprehensive study. The whole nature of the solutions may change as we vary the longitudinal and transverse relaxation times of the lasing transition, the cavity round-trip time, and the cavity losses. In addition, the choice of initial conditions is important since multistable solutions are possible.[4] Furthermore, the validity of the approximations used in the numerical analysis becomes unreliable in the vicinity of the critical points where the system becomes unstable. In these regions, the noise in the system becomes important in determining the dynamics.[5]

An alternative approach is to carry out laser experiments looking for new modes of operation. There have been a number of such experiments over the years. As early as 1968, Bass *et al.* reported apparent stable two-frequency operation of an argon-ion laser.[6] More recently, there have been a number of studies of instabilities in inhomogeneously broadened lasers.[7–9] In a beautiful set of experiments, Maeda and Abraham and Gioggia and Abraham have observed self-pulsing, quasi-periodic, and apparently chaotic fluctuations in the intensity output of a cw-pumped laser.[10] However, the presence of inhomogeneous broadening, spatial hole burning, pump fluctuations, etc., greatly complicates and enriches the possible states of the system.[11,12] By following analogies with other driven nonlinear systems, one is left wondering whether there exist instabilities and new types of stable states in the simplest type of laser, the "ideal" homogeneously broadened laser.[13]

In a recent publication, we reported the observation of instabilities and a series of new stable states of operation of a cw-pumped, homogeneously broadened, ring dye laser.[14] This observation showed that when such a laser is pumped far enough above threshold it will oscillate simultaneously at two frequencies symmetrically displaced from line center.

In the present paper, we briefly review the results of this experiment in Section 2. In Section 3, we calculate the saturated gain for an ideal homogeneously broadened laser oscillating at two optical frequencies symmetrically detuned about the center frequency of the gain profile. We show that

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this gain curve exhibits a series of maxima for frequency separations equal to the Rabi frequency and subharmonics of it. Finally, we include the effects of dispersive loss and compare the results with experimental observations.

2. REVIEW OF EXPERIMENTAL RESULTS

Hillman *et al.*[14] studied the operating characteristics of a Rhodamine 6G ring dye laser pumped by a cw argon-ion laser. The details of the cavity design are given in Ref. 14. For our purposes, we need only note a few facts about the laser. First, all the mirrors are broadband high reflectors, so the Q of the cavity is about 300. This means that 2 W of pump-laser power produces approximately 80 W of circulating dye-laser power in the cavity, which corresponds to a 15% pump-conversion efficiency. At the beam waist in the dye jet, this results in approximately 50 MW/cm^2 of the dye-laser intensity. This intensity generates a Rabi frequency in the dye transition as large 10^{13} Hz, which corresponds to a wavelength splitting of more than 50 Å. Second, a Brewster-angle prism is placed in the cavity to produce a slight frequency-dependent loss centered at the peak of the gain curve.

In the experiment, two quantities were measured as the pump power was increased: the intracavity power and the spectrum of the dye-laser radiation. Figure 1 shows the experimentally measured dependence of intracavity power on the argon-pump power. At point A, the laser reaches threshold and begins to oscillate at a single frequency. At point B, the laser reaches a second threshold at which it abruptly switches to two-frequency operation at point C. If the pump power is then decreased slightly, a hysteresis loop is seen in the switch back to single-frequency operation. When the pump-laser power is increased further, the laser oscillates stably at two frequencies until a third threshold is reached at point D. At this point, there is again an abrupt switch in the state of the laser; the cavity power jumps to point F. There is a much larger hysteresis loop about this threshold. In the region from F to G, the laser again operates stably with a two-frequency output, but the separation between the two frequency components is less than that at point D.

In Fig. 2(a), the spectral output of the laser is plotted as a function of the intracavity power. The range of powers corresponds to the region from C to D in Fig. 1. The laser output is two-frequency throughout this region, and the observed wavelengths fall accurately on a parabola. The data are noisier for the region E–G, but, as can be seen in Fig. 2(b), the spectral splitting again appears to fall on a parabola as a function of the intracavity power.

These results give us the clues that we need to narrow our search for new stable solutions to the laser equations. We see that the single-mode solutions are unstable far above threshold, as was predicted by the linear stability analysis, and we see that the new stable states are bichromatic. As the sidebands grow, they quench the original lasing frequency and then exist stably by themselves. The separation between the two frequency components follows a parabolic dependence as a function of intracavity power, which we would expect if the splitting were due to the Rabi oscillations. The occurrence of Rabi oscillations points out the fact that a fully coherent treatment of the gain medium is required since Rabi oscillations do not occur in rate equations.

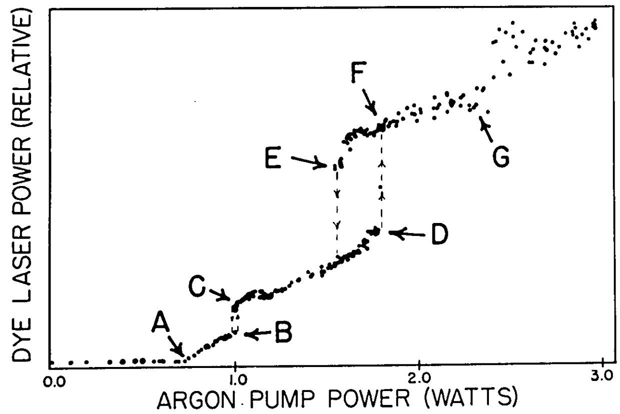

Fig. 1. Experimentally measured dependence of intracavity power on the argon-pump power. Letters indicate the various thresholds of instability; see corresponding points in Fig. 2. Single-frequency operation occurs only between points A (lasing threshold) and B.

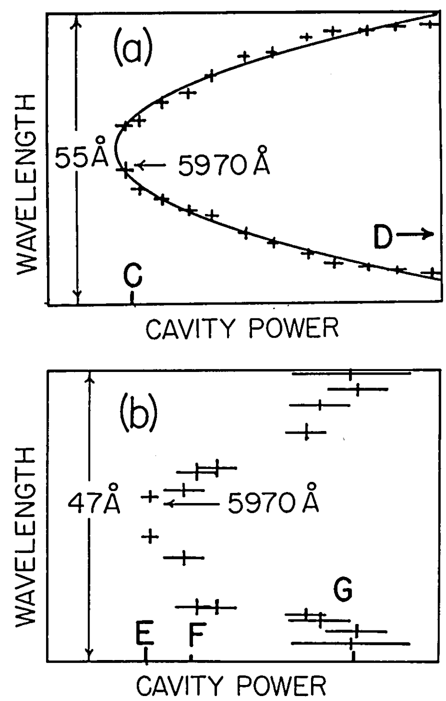

Fig. 2. Spectral output of the dye laser as a function of cavity power. (a) The pairs of operation frequencies on the C, D branch have a parabolic dependence on the cavity power. (b) The pairs of operation frequencies on the E–G branch appear to fall on another parabola.

In the experiment, the observed spectral splittings (~50 Å) are of the order of the $1/T_2$ homogeneous linewidth of Rhodamine 6G. However, in comparison, the spacing of adjacent longitudinal cavity modes is only 1.2 GHz for the 25-cm ring cavity, which is beyond the resolution of the spectrometer used and some 5000 times finer than the observed structure. In addition, the spectral selectivity and the mechanical stability of the cavity are not sufficient to keep the laser operating in two particular longitudinal modes for the time required to scan the entire spectrum; yet the spectrum is quite stable in form. The large difference in scale between the observed structure and the cavity-mode separation, and the insensitivity of the structure to hopping between closely spaced cavity modes, suggests that the cavity-mode spectrum plays no significant role and therefore can be treated as a continuum for the range of parameters studied here. Rather, it seems that only the atomic dynamics, which are characterized by the Rabi frequency, play the dominant role in determining the laser's state of operation.

The data provide us with an additional clue that had not been previously predicted. The bichromatic state itself eventually becomes unstable as the intracavity power is increased. After a third threshold, another stable bichromatic state occurs. The theory must predict more than one possible bichromatic state.

3. THEORY

The experimental clues discussed in Section 2 suggest that we should investigate the laser equations by assuming that only two strong fields symmetrically displaced in frequency about the line center are oscillating. This is illustrated in Fig. 3. Such a field may be written as

$$E(t) = 2\mathscr{E}_1[\cos(\omega_0 + \delta\omega)t + \cos(\omega_0 - \delta\omega)t]$$
$$= 4\mathscr{E}_1 \cos \delta\omega t \cos \omega_0 t. \qquad (1)$$

Here, we have taken $\mathscr{E}_1$ to be a time-independent amplitude, ω_0 to be the gain-center frequency, and $\delta\omega$ to be the frequency displacement from gain center. Note that the field may be written equivalently as a sinusoidally amplitude-modulated field at frequency $\delta\omega$. We now calculate the gain as a function of $\delta\omega$ seen by this bichromatic field interacting with the saturable medium.

The gain medium is modeled by a two-level atomic resonance that is described by the optical Bloch equations

$$\dot{v} = -v/T_2 + 2\kappa\mathscr{E}_1 w \cos \delta\omega t,$$
$$\dot{w} = -(w - w_{\rm eq})/T_1 - 2\kappa\mathscr{E}_1 v \cos \delta\omega t. \qquad (2)$$

The notation is that of the text by Allen and Eberly,[15] in which v is the atomic polarization in the frame oscillating as $\cos \omega_0 t$, w is the inversion, $\kappa \equiv 2d/\hbar$ is the coupling constant between the dipole moment whose matrix element is d and the electric field, T_1 and T_2 are, respectively, the longitudinal and transverse relaxation times, and $w_{\rm eq} > 0$ is the equilibrium value to which the inversion relaxes in the absence of a resonant field.

The harmonic response of the atom to this bichromatic field may be obtained by making a Floquet expansion of v and w:

$$v(t) = \sum_{n=-\infty}^{\infty} \tilde{v}_n \exp(in\delta\omega t),$$
$$w(t) = \sum_{n=-\infty}^{\infty} \tilde{w}_n \exp(in\delta\omega t). \qquad (3)$$

By substituting these expansions into Eqs. (2) we obtain the recurrence relations

$$(1 + in\delta\omega T_2)\tilde{v}_n = \kappa\mathscr{E}_1 T_2(\tilde{w}_{n+1} + \tilde{w}_{n-1}),$$
$$(1 + in\delta\omega T_1)\tilde{w}_n = w_{\rm eq}\delta_{n,0} - \kappa\mathscr{E}_1 T_1(\tilde{v}_{n+1} + \tilde{v}_{n-1}). \qquad (4)$$

We can solve these relations to obtain a continued-fraction solution for the components of v or w oscillating at any harmonic of the modulation frequency $\delta\omega$.

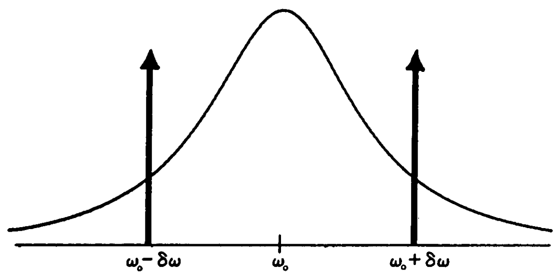

Fig. 3. The assumed bichromatic operating state of the laser. The total laser field is made up of two strong-field components of equal amplitude that are symmetrically displaced from the gain center.

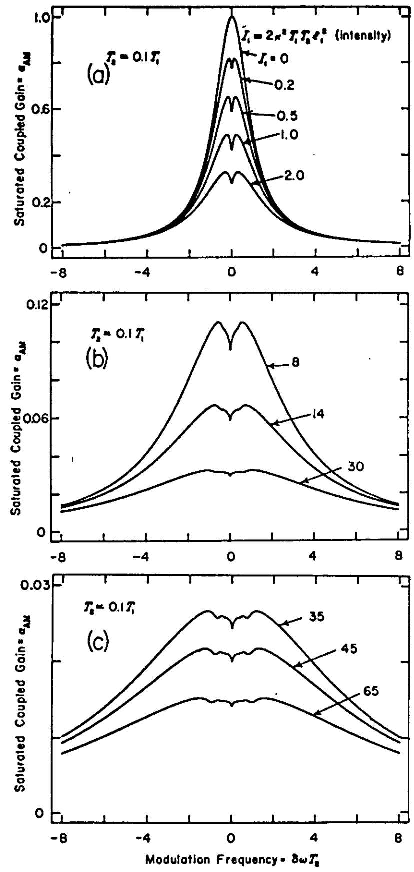

Fig. 4. The saturated gain of the total bichromatic field at various intensities as a function of the modulation frequency. (a) At low intensity, a spectral hole of width $\sim 1/T_1$ appears in the gain profile. The maximum gain for a bichromatic field occurs for nonzero modulation. (b) At higher intensity, the gain curve saturates, and structure begins to appear about the hole at line center. (c) This structure becomes more complex at higher intensities. Additional maxima appear in the gain profile at several different modulation frequencies.

The time-averaged gain $\alpha_{\rm AM}$ seen by the bichromatic field is proportional to $\tilde{v}_1$:

$$\alpha_{\rm AM} = \frac{\alpha}{\kappa T_2 \mathscr{E}_1} {\rm Re}\{\tilde{v}_1\}$$
$$= \frac{\alpha\, {\rm Re}\{S\}}{1 + 2I_1\, {\rm Re}\{S\}}. \qquad (5)$$

In Eq. (5), α is the unsaturated gain that would be seen by a monochromatic field at line center, and $I_1 \equiv 2\kappa^2 T_1 T_2 \mathscr{E}_1{}^2$ is the dimensionless intensity of the bichromatic field. The con-

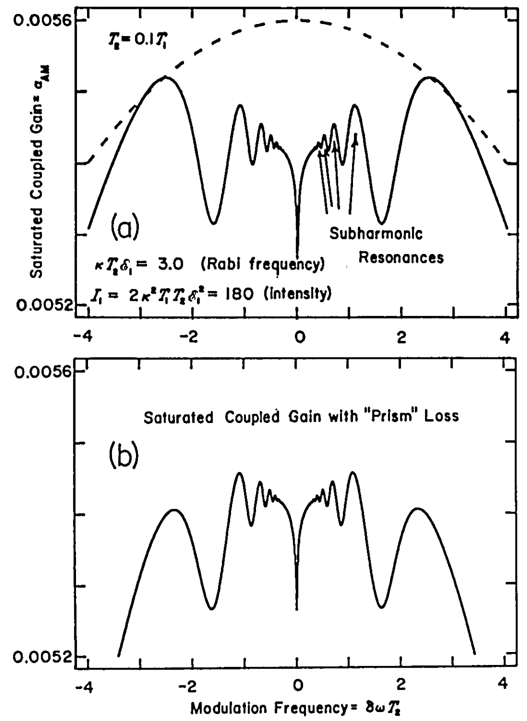

Fig. 5. (a) Expanded view of the structure in the gain profile at high laser intensity. The largest gain for a bichromatic field occurs when the modulation frequency is approximately equal to the Rabi frequency. Additional maxima occur at subharmonics of the Rabi frequency. The dashed curve represents dispersive losses typical of a prism. (b) The net saturated gain for a laser operating at two frequencies and containing a prism. The first subharmonic resonance has greater net gain than the fundamental resonance. The laser therefore operates more efficiently at this subharmonic resonance.

tinued fraction S may be written as

$$S = \cfrac{1}{B_1 + \cfrac{I_1}{B_2 + \cfrac{I_1}{B_3 + \ddots}}}\,,$$

where

$$B_n \equiv \begin{cases} 1 + in\delta\omega T_2, & n \text{ odd} \\ 1 + in\delta\omega T_1, & n \text{ even} \end{cases}. \qquad (6)$$

Similar continued-fraction solutions describing the atomic response to multicomponent fields have been obtained by others.[16] In Fig. 4, this expression is plotted as a function of the modulation frequency for various intensities I_1 for the case of $T_2/T_1 = 0.1$. At low intensities, the gain curve is a simple Lorentzian with maximum gain for a modulation frequency of zero. As the intensity is increased, saturation occurs, and a spectral hole appears at line center. The highest gain is experienced by a bichromatic field with a nonzero modulation frequency. As the intensity of the bichromatic field increases further, the whole curve saturates, and the spectral hole broadens. In Fig. 4(b), we see that at yet higher intensities new features appear. Additional structure appears within the hole. In Fig. 4(c), curves are shown for the case of high intensities at which the additional structure becomes increasingly complex.

In Fig. 5(a) the central portion of the gain curve is expanded to show the detailed structure. The curve is labeled with the Rabi frequency as well as the intensity.[17] It can be seen that there is a series of maxima in the gain curve. The outermost maximum corresponds to a modulation frequency approximately equal to the Rabi frequency; the second maximum corresponds to a modulation frequency approximately equal to one half of the Rabi frequency. Several other subharmonics of the Rabi frequency also produce maxima in the gain curve. At the center, there is a hole of width approximately equal to $1/T_1$.

The nature of these resonances is easily understood. The inversion is coherently driven to oscillate at the Rabi frequency. Maximum gain occurs when the field is modulated at the same frequency as this oscillation. A secondary maximum occurs when the field is modulated at one half of the rate at which the inversion oscillates. Additional resonances occur at the other subharmonics of the Rabi frequency. These resonances are analogous to those that occur when a swing is pushed once each round trip, once every second round trip, once every third round trip, etc. Because of damping, only a finite number of the subharmonic resonances can occur; the number depends on the level of excitation. The damping also slightly modifies the exact subharmonic relationship. A related set of resonances for the case of an absorber was predicted by Shirley[18] and Thomann[19] and was observed in an experiment by Thomann.[19] This substructure in the absorption spectrum of a two-level system has also been observed with Zeeman-split levels resonantly excited by radio-frequency fields.[20]

The experimental results can now be understood. At low intensities, the field is too weak to overcome the damping and produce Rabi oscillations. Single-frequency operation then offers the highest gain. As the pumping is increased, the field grows sufficiently strong that Rabi oscillations induce side-mode gain. In this case, a new type of operation in which the field is amplitude modulated at a frequency just equal to the

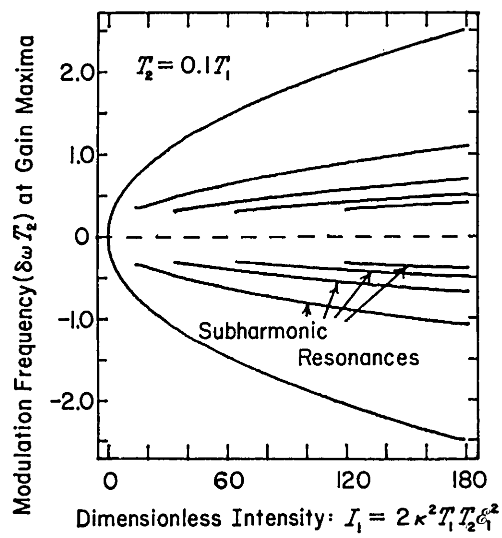

Fig. 6. Location of maxima in the gain profile as a function of intensity. The outer curve is a parabola that corresponds to modulation at the Rabi frequency. The other curves are portions of parabolas that correspond to modulation at subharmonics of the Rabi frequency. The particular intensity at which each of the subharmonic resonances first appears is determined by the relaxation times of the gain medium.

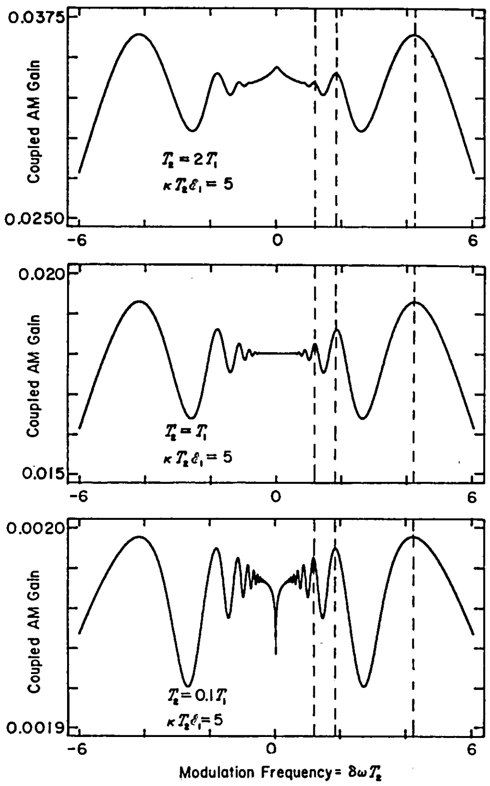

Fig. 7. Comparison of gain profiles for three ratios of the damping times T_1 and T_2 with a fixed Rabi frequency. Note that the maxima in the gain profiles are identical for each of the cases. Only the central structure within the subharmonic resonances is changed by changing the ratio of T_1 to T_2.

Rabi frequency has higher gain. The laser then becomes unstable and switches to the state with higher gain.

If we increase the field intensity further, the Rabi frequency increases, and the two frequency components of the field are pushed farther and farther from line center. At a given level of excitation, the gain is always greater for a modulation frequency equal to the Rabi frequency; therefore, one would not expect further instabilities to develop. However, an important feature of the laser experiment has been omitted. A dispersive element, namely, a prism was included in the cavity. When the lasing frequencies are far enough from line center, they suffer additional losses. Such losses can be included in our theory by multiplying the gain at each frequency by a corresponding loss. If we model this loss by a parabola, as is shown by the dashed curve in Fig. 5(a), we get the modified gain curve shown in Fig. 5(b). The gain at the Rabi frequency is suppressed, so the gain at the first subharmonic is now highest. If the intensity is increased still further, the first subharmonic will be pushed farther out, increasing its loss. The second subharmonic will then eventually have the highest gain. In this way, as the intensity is increased, the laser will pass through a series of thresholds at which the various subharmonics in turn become the states with highest net gain. The laser will become unstable at each threshold and jump discontinuously to the state with highest gain.

In Fig. 6, the locations of the various maxima in the gain curve are plotted as a function of the intensity. Each of the curves is a parabola, or a portion of a parabola, which corresponds to a Rabi-frequency dependence. At low intensities, there is a maximum only at the Rabi frequency. As the intensity is increased, successive subharmonics reach thresholds at which they begin to produce local maxima. These curves agree qualitatively with the observations of Hillman *et al.*[14]

In all our examples thus far we have taken $T_2/T_1 = 0.1$. The ratio of these damping times varies greatly from one medium to the next; the effect of this variation is illustrated in Fig. 7. For a fixed value of the Rabi frequency, $\kappa T_2 \mathscr{E}_1 = 5$, we illustrate three cases. At the top of Fig. 7 is the case of purely radiative broadening, $T_2/T_1 = 2$; in the center is the case of hard collisional damping, $T_2/T_1 = 1$; and at the bottom is the case of collisional dephasing, $T_2/T_1 = 0.1$. In each case, the locations of the maxima are the same, so the predicted bichromatic states are identical. The structure at the center, inside all the subharmonic resonances, is different in each case. The stability of these bichromatic states may depend on the ratio of the damping constants.

4. DISCUSSION

This paper represents the first reported analysis of a new regime in which a homogeneously broadened laser can operate. Led by our experimental observations, we have analyzed the saturated gain of a laser for the case of simultaneous oscillation at two optical frequencies. Unlike previous calculations on multifrequency lasers, the present analysis allowed the two fields to be arbitrarily strong and treated the gain medium not by population rate equations but by the coherent Bloch equations. We have found that the gain profile of this bichromatic field exhibits a series of resonant maxima that occur when the modulation frequency of the field is equal to the Rabi frequency or one of its subharmonics. These resonances in the gain profile explain both the parabolic power dependence of the wavelength splitting and the additional bichromatic states that we have observed in our ring dye laser. This mechanism is perhaps also responsible for the power dependence of the modulation that has been observed in semiconductor lasers[21] and argon-ion lasers.[6]

Although we have presented results only for the case of purely homogeneous broadening, resonances induced by Rabi oscillations will be present in all lasers, including those with inhomogeneous broadening and standing-wave cavities. In a gas laser with a standing-wave cavity, a moving molecule will see a field that is amplitude modulated at a frequency proportional to its velocity. Therefore there are resonances when the modulation frequency of the field seen by a molecule is equal to its Rabi frequency or a subharmonic of the Rabi frequency. In fact, the research by Feldman and Feld[22] on the theory of a single-mode gas laser clearly exhibits this structure within the Bennett dip (see Fig. 8 of Ref. 22). These subharmonic resonances may also provide the necessary mechanism that induces the period-doubling bifurcations and the eventual chaotic behavior that has been observed in inhomogeneously broadened lasers.[23]

In this paper, we have discussed a series of states in which a cw homogeneously broadened laser can operate that are neither single mode nor mode locked in the conventional sense. Our analysis of such a laser is incomplete; however, the initial results are fascinating. Whether this series leads eventually to normal mode locking or to a chaotic state, we cannot say. We are currently extending the results and investigating the stability of these bichromatic solutions. In conclusion, it is clear from these initial studies that lasers operating far above threshold possess a number of possible states whose properties are not yet well understood.

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* Present address, Kodak Research Laboratory, Rochester, New York 14650.

† Present address, Allied Corporation, Mt. Bethel, New Jersey 09060.

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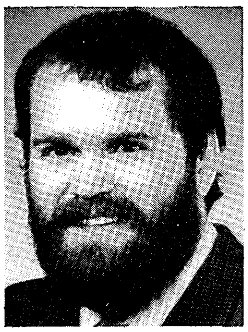

Lloyd W. Hillman was born in Ann Arbor, Michigan in 1955. He received the bachelor's degree in engineering physics from the University of Arizona in 1976 and a Ph.D. degree from the Institute of Optics at the University of Rochester in 1984. His thesis work focused on optical bistability, laser instabilities, and the interaction of modulated fields with matter. He is member of the Optical Society of America and is employed at the Kodak Research Laboratory in Rochester.

Jerzy Krasinski

Jerzy Krasinski received the M.S. degree in physics in 1966 and the Ph.D. degree in 1973 from the University of Warsaw in Poland. His thesis work was concerned with photon statistics and nonlinear absorption. In 1973 he became an *adjunkt* (equivalent to an assistant professor) and in 1977 a *docent* (equivalent to tenured associate professor) and head of the Optics Department of the University of Warsaw. He also had visiting positions at Indiana University, Max Planck Institute in Garching bei München, Colgate University, and the University of Rochester (twice). Since 1982, he has lived in the United States, and in 1984 he became senior research physicist at Allied Corporation. His research has involved nonlinear optics, laser spectroscopy, new laser media, and development of new laser systems.

Karl Koch

Karl Koch is a graduate student at the Institute of Optics at the University of Rochester. He was born in New London, Connecticut in 1960. He received the bachelor's degree in physics from San Diego State University in 1982. His research interests include laser instabilities, optical bistability, and other subjects in quantum optics. He is a member of the Optical Society of America and the American Physical Society.

C. R. Stroud, Jr.

C. R. Stroud, Jr., is Professor of Optics at the University of Rochester. He was born in Owensboro, Kentucky, and received the bachelor's degree in mathematics and physics from Centre College of Kentucky and the Ph.D. degree with a thesis in theoretical physics from Washington University. He has been a member of the faculty of the Institute of Optics since 1969, working in experimental and theoretical quantum optics. He is a fellow of both the American Physical Society and the Optical Society of America.