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

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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 center frequency of the gain profile. We show that 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
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.\textsuperscript{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\textsuperscript{2} of the dye-laser intensity. This intensity generates a Rabi frequency in the dye transition as large 10\textsuperscript{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-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.
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

\[ E(t) = 2 \mathcal{E}_1 \left[ \cos(\omega_0 + \delta \omega)t + \cos(\omega_0 - \delta \omega)t \right] = 4 \mathcal{E}_1 \cos \delta \omega t \cos \omega_0 t. \]  

(1)

Here, we have taken \( \mathcal{E}_1 \) to be a time-independent amplitude, \( \omega_0 \) 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

\[ \dot{v} = -v/T_2 + 2\kappa \mathcal{E}_1 \cos \delta \omega t, \]
\[ \dot{w} = -(w - w_{eq})/T_1 - 2\kappa \mathcal{E}_1 \cos \delta \omega t. \]  

(2)

The notation is that of the text by Allen and Eberly,\(^1\) in which \( v \) is the atomic polarization in the frame oscillating as \( \cos \omega_0 t \), and \( w \) is the inversion, \( \kappa = 2d/h \) 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 \( w_{eq} > 0 \) 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 \( w \):

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

(3)

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

\[ (1 + in \delta \omega T_2)\tilde{v}_n = \kappa \mathcal{E}_1 T_2(\tilde{w}_{n+1} + \tilde{w}_{n-1}), \]
\[ (1 + in \delta \omega T_1)\tilde{w}_n = w_{eq} \delta_{n0} - \kappa \mathcal{E}_1 T_1(\tilde{v}_{n+1} + \tilde{v}_{n-1}). \]  

(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 \( \delta \omega \).

The time-averaged gain \( \alpha_{AM} \) seen by the bichromatic field is proportional to \( \tilde{v}_1^2 \):

\[ \alpha_{AM} = \frac{\alpha}{\kappa T_2^2 \mathcal{E}_1} \text{Re}[\tilde{v}_1] = \frac{\alpha \text{Re}[S]}{1 + 2I_1 \text{Re}[S]}. \]  

(5)

In Eq. (5), \( \alpha \) is the unsaturated gain that would be seen by a monochromatic field at line center, and \( I_1 = 2k^2T_1^2T_2^2 \mathcal{E}_1^2 \) is the dimensionless intensity of the bichromatic field. The con-
Increasingly complex. Intensities at which the additional structure becomes in-the hole. In Fig. 4(c), curves are shown for the case of high new features appear. Additional structure appears within broadens. In Fig. 4(b), we see that at yet higher intensities further, the whole curve saturates, and the spectral hole frequency. As the intensity of the bichromatic field increases experienced by a bichromatic field with a nonzero modulation of zero. As the intensity is increased, saturation occurs, and the largest gain is Lorentzian with maximum gain for a modulation frequency of 

\[ \frac{1}{T_1} = \frac{1}{T_1} \] 

Similar continued-fraction solutions describing the atomic response to multicomponent fields have been obtained by others. 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. 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 and Thomann and was observed in an experiment by Thomann. 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.

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 sidemode gain. In this case, a new type of operation in which the field is amplitude modulated at a frequency just equal to the
The laser will become unstable at each threshold and jump harmonics in turn become the states with highest net gain. If the intensity is increased still further, the first subharmonic is suppressed, so the gain at the first subharmonic is now equal to its Rabi frequency or a subharmonic of the Rabi frequency; therefore, one would not 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 Feld22 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 in-

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, $\kappa T_2 \delta_1 = 5$, 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 lasers21 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 Feld22 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

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vestigating 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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REFERENCES

17. Although a Rabi frequency is usually defined with a constant field, we have associated an equivalent Rabi frequency whose value is equal to the time-averaged value of the root-mean-square modulated field. By this definition, the Rabi frequency is still related to the average rate of excitation.
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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.

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