Observation of Higher Order Dynamical States of a Homogeneously Broadened Laser

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A series of new stable operating states of a homogeneously broadened laser have been observed. As the pump power is varied, thresholds are reached where the power and spectrum of the laser change discontinuously. In these states the laser simultaneously oscillates at two frequencies whose separation is given by the Rabi frequency or one of its subharmonics. A theoretical explanation is given which includes field-induced structure within the homogeneous line.

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Recent work has shown that the basic physics of cw-pumped lasers is richer than was previously thought.^{1,2} A number of dynamical instabilities that begin with spontaneous pulsations³ and may eventually lead to chaotic operation have been observed in inhomogeneously broadened⁴ and actively modulated lasers.⁵ Recent observations of fieldinduced structures within a homogeneously broadened line⁶ suggest that the homogeneously broadened laser can also show similar behavior.

We have observed a series of instabilities in a homogeneously broadened dye laser. Our laser consists of a simple ring cavity with no modulators or saturable absorbers. At high intracavity power, critical transitions occur whereby, instead of singlefrequency oscillation, the laser oscillates at two frequencies whose spectral separation is equal to the Rabi frequency or a subharmonic of the Rabi frequency. We can observe stable operating regions of the laser, separated by well-defined thresholds. Wide hysteresis loops as well as bistable operation of the laser are observed as the pump power is changed. These higher-order dynamic states, intermediate between those describing cw and either mode-locked or chaotic operation, occur because the intense intracavity field actively modifies the homogeneous gain profile.

Questions about the stability of a laser's output figured prominently in the earliest literature starting with the operation of the first ruby laser by Maiman.⁷ Effects discussed in the early literature that can spoil single-frequency operation and lead to pulsations in the output power include relaxation oscillations,⁸ spectral hole burning in inhomogeneously broadened media,⁹ and spatial hole burning in standing-wave cavities.¹⁰ For the case of a cwpumped, homogeneously broadened, unidirectional ring laser, none of these can occur. However, it is found in the laboratory that the achievement of

stable single-mode operation of a laser at high output power requires considerable engineering detail, even though conventional rate equations predict that only one mode can oscillate in the ideal homogeneous case. Early work by Haken¹¹ and Risken, Schmid, and Weidlich¹² reveals an interesting result-at high photon density, amplitude fluctuations in a single mode laser can become unstable. Risken and Nummedal¹³ present a simple semiclassical treatment of this instability. The strong intracavity field modifies the gain profile such that cavity-field components detuned from the oscillation frequency of the laser by the Rabi frequency experience gain. When this parametric gain exceeds the threshold value, the single-frequency laser becomes unstable to the growth of these new frequency components. This parametric coupling of the strong laser field to its frequency sidebands is intrinsic to all lasers. There have been a number of other studies on various aspects of this instability^{14,15} including the effect of inhomogeneous broadening.^{16, 17} In an inhomogeneously broadened laser, several processes contribute to instability; however, in a homogeneously broadened ring laser only the intrinsic instability discussed by Risken and Nummedal¹³ can occur. Our experiment is performed on such a laser.

A rhodamine 6G ring dye laser pumped by a cw argon-ion laser is used in our experiment. The high-Q ring cavity of length 25 cm is constructed with broadband highly reflecting mirrors and a weakly dispersive prism. The cavity losses are so low that only 2 W of pump power generate more than 85 W of circulating dye-laser power, corresponding to an intensity of greater than 50 MW/cm² in the dye jet. The circulating power is determined by a measurement of the intensity of Rayleigh scattering of the intracavity laser beam.¹⁸ The dye laser is adjusted so that it operates at the center of the dye gain curve in the lowest-order (Gaussian) transverse mode. The spectral output of the laser is directly measured with a one-meter spectrometer with a vidicon at the back focal plane.

Figure 1 shows the dependence of the dye laser power on the argon pump power. As the pump power is increased from zero, the dye laser first reaches threshold (marked A in Fig. 1), and operates at gain center, its power increasing linearly with pump power. Increasing the pump power further, we find a second threshold at which a discontinuous increase in the power of the dye laser occurs. At this point, the dye laser switches from (B)operation at a single frequency at gain center to (C) operation simulataneously at two frequencies symmetrically displaced from gain center. The transition from a one-frequency laser to a twofrequency laser is analogous to a first-order phase transition. Figure 1 shows that this instability is bistable; a small hysteresis loop occurs about the critical power.

As the pump power is further increased, we again find a nearly linear dependence of the dye laser power on the pump power (from C to D in Fig. 1). However, the spectral output displays a strong power dependence as illustrated in the left column of Fig. 2. The two lasing wavelengths split further apart as the cavity power increases. Splittings as large as 75 Å are observed. At the top of the left column of Fig. 2, we plot the wavelengths of the two lasing frequencies versus the relative power in the dye laser. We find that the dependence of the wavelength splitting on the cavity power is fitted closely by a parabola. This dependence is consistent with that produced by Rabi oscillations, which



FIG. 1. Dye-laser power vs pump power. Special points in the figure are (A) the lasing threshold; (B) second threshold; and (D,G) higher thresholds. Hysteresis loops occur at B-C and D-E-F.

strongly couple the two fields.

At still higher pump powers, another discontinuous jump in the dye-laser power occurs. This instability is preceded by the growth of a mode at the center of the gain curve. This mode rapidly grows, then bifurcates with the simultaneous extinction of the outer two modes. This higher-order instability exhibits a large hysteresis loop (D, E, F in Fig. 1). The spectral output of the dye laser along the branch F, E, G is illustrated in the right column of Fig. 2. Although the data are rather noisy, a parabolic dependence of the splitting with the cavity power is again observed. This dependence indicates that the two strong fields are coupled by atomic oscillations at a submultiple of the Rabi frequency. Beyond G, further power jumps (including negative jumps), hysteresis, and spectral bifurcations occur, leading to a seemingly chaotic state.

The threshold for the instability in a homogeneously broadened laser is predicted by a linear stability analysis.¹³ In this analysis, infinitesimal perturbations about the stationary single-frequency solution experience exponential growth; therefore, the initial state is unstable. The field associated with the unstable perturbation corresponds to the bichromatic linear combination of two phase-locked, single-frequency probe fields that are symmetrically



FIG. 2. Power dependence of dye laser spectrum. The letters indicate corresponding points in Fig. 1. In the upper left the wavelength splitting of the spectrum as a function of laser power is compared with a parabola.

detuned from the oscillation frequency of the laser by the Rabi frequency. The single-frequency laser becomes unstable when the parametric gain of this bichromatic "natural mode"¹⁵ of the probe field exceeds the round-trip cavity loss threshold. Once this second threshold is crossed, the sidebands become finite and the perturbative analysis is no longer valid.

To investigate the dynamic state of the laser beyond the second threshold, we have solved analytically the problem of three AM-phase-locked fields of arbitrary strength interacting with a twolevel atomic medium. The details of this solution will appear elsewhere. Several of the main results of this solution explain observations made in our experiment. At the onset of the instability, the two side modes grow and mutually support each other while they quench the gain of the center; therefore, the single-frequency laser bifurcates to a twofrequency laser. This bifurcation is accompanied by a discontinuous increase in the cavity power. The two-mode solution exhibits bistability; it remains stable with decrease of the pump power below the second threshold value.

In the special case in which the two homogeneous lifetimes are equal, $T_1 = T_2$, the general solution for the coupled gain of two symmetrically detuned fields of equal amplitude E_1 , reduces to the equation

$$\alpha_{\rm AM} = \alpha \sum_{n=1}^{\infty} \left(\frac{n \, \delta \omega}{\kappa \, \mathcal{C}_1} \right)^2 \frac{J_n^2 (2 \kappa \mathcal{C}_1 / \delta \omega)}{1 + (n \, \delta \omega \, T_2)^2},\tag{1}$$

where α is the unsaturated gain coefficient at line center, $\kappa \mathscr{C}_1$ is the Rabi frequency, $\delta \omega$ is the detuning of the two field components from line center, and J_n are Bessel functions of order *n*. The frequency separation of the two oscillating fields is determined by the localized gain maxima of the coupled solution. At low power, the coupled solution exhibits only one gain maximum located at $\delta \omega$ equal to the Rabi frequency. However, at higher powers we find that localized gain maxima appear at subharmonics of the Rabi frequency, as illustrated in Fig. 3. Similar sub-Rabi-frequency resonant structure is found in other work with strong fields interacting with atomic transitions.¹⁹ A stability analysis of the two-frequency solutions shows that only over a limited range of detuning about each maxima are the solutions stable against the growth of the central mode. Because of the frequency pulling effects of the weakly dispersive prism, the widely split frequencies on the first branch in our experiment reach an instability point at which the laser frequency jumps to one of the inner submaxima.



FIG. 3. Theoretical gain curve for operation at two frequencies symmetrically detuned from line center. The cross-hatching indicates regions of stable two-frequency operation for a laser at either the Rabi frequency or submultiples of it.

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In summary, we have observed a series of instabilities in the intracavity power and spectral output of a homogeneously broadened ring laser. These instabilities are due to the response of individual atoms interacting with a strong field, and therefore are termed "intrinsic" by Risken and Nummedal.¹³ Two-frequency operation with a spectral separation equal to the Rabi frequency is observed when the laser operates above the so-called second threshold. Above a third threshold, the separation is a subharmonic of the Rabi frequency. These results are consistent with new theoretical predictions for the saturated gain of a laser operating at two frequencies and provide a mechanism for the subharmonic bifurcations that have been observed in inhomogeneously broadened lasers.¹

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¹R. S. Gioggia and N. B. Abraham, Phys. Rev. Lett. **51**, 650 (1983).

²L. W. Casperson, Phys. Rev. A **21**, 911 (1980), and **23**, 248 (1981).

³K. R. Manes and A. E. Siegman, Phys. Rev. A 4, 373 (1971); Casperson and A. Yariv, IEEE J. Quantum Electron. 8, 69 (1972); J. Bentley and N. B. Abraham, Opt. Commun. 41, 52 (1982).

⁴M. Maeda and N. B. Abraham, Phys. Rev. A **26**, 3395 (1982); R. S. Gioggia and N. B. Abraham, Opt. Commun. **47**, 278 (1983).

⁵F. T. Arecchi, R. Meucci, G. Puccioni, and J. Tedicce, Phys. Rev. Lett. **49**, 1217 (1982).

⁶L. W. Hillman, R. W. Boyd, J. Krasinski, and C. R. Stroud, Jr., Opt. Commun. **45**, 416 (1983); M. Malcuit, R. W. Boyd, L. W. Hillman, J. Krasinski, and C. R. Stroud, Jr., J. Opt. Soc. Am. B **1**, 73 (1984).

⁷T. H. Maiman, Phys. Rev. **123**, 1151 (1961).

⁸H. Statz and D. DeMars, in *Quantum Electronics*, edited by C. H. Townes (Columbia Univ. Press, New York, 1960), p. 530.

⁹W. R. Bennett, Jr., Phys. Rev. 126, 580 (1962).

¹⁰W. E. Lamb, Jr., Phys. Rev. **134**, A1428 (1964).

¹¹H. Haken, Z. Phys. **190**, 327 (1966).

¹²H. Risken, C. Schmid, and W. Weidlich, Z. Phys. **194**, 337 (1966).

¹³H. Risken and K. Nummedal, Phys. Lett. **26A**, 275 (1968), and J. Appl. Phys. **39**, 4662 (1968).

¹⁴R. Graham and H. Haken, Z. Phys. 213, 420 (1968);

C. L. Tang and H. Statz, J. Appl. Phys. 39, 31 (1968);

H. Haken and H. Ohno, Opt. Commun. 16, 205 (1976),

and 26, 117 (1978); P. Mandel and H. Zeghlache, Opt. Commun. 47, 146 (1983).

¹⁵L. W. Hillman, R. W. Boyd, and C. R. Stroud, Jr., Opt. Lett. **7**, 426 (1982).

¹⁶S. T. Hendow and M. Sargent, III, Opt. Commun. **40**, 385 (1982), and **43**, 59 (1982); L. A. Lugiato, L. M. Narducci, D. K. Bandy, and N. B. Abraham, Opt. Commun. **46**, 115 (1983).

¹⁷M. L. Minden and L. W. Casperson, IEEE J. Quantum Electron. **18**, 1952 (1982); P. Mandel, Opt. Commun. **44**, 400 (1983), and **45**, 269 (1983).

¹⁸L. W. Hillman, J. Krasiński, J. A. Yeazell, and C. R. Stroud, Jr., Appl. Opt. **22**, 3474 (1983).

¹⁹B. J. Feldman and M. S. Feld, Phys. Rev. A **1**, 1375 (1970); N. Tsukada and T. Nakayama, Phys. Rev. A **25**, 964 (1982); H. J. Carmichael and G. P. Agrawal, in *Optical Bistability*, edited by C. M. Bowden, M. Ciftan, and H. R. Robl (Plenum, New York, 1981), p. 237.