Nonlinear dynamics of a multimode dye laser with an adjustable resonator dispersion and implications for the sensitivity of in-resonator laser spectroscopy

S. E. Vinogradov, A. A. Kachanov, S. A. Kovalenko, and É. A. Sviridenkov
P. N. Lebedev Physics Institute, Russian Academy of Sciences, 117924, Moscow

(Submitted 9 April 1992)
Pis'ma Zh. Eksp. Teor. Fiz. **55**, No. 10, 560–563 (25 May 1992)

The effect of nonlinear mode interactions has been eliminated through a cancellation of the resonator dispersion of a wide-band ring laser. A sensitivity better than 10^{-10} cm^{-1} has been achieved in in-resonator laser spectroscopy.

The operation of multimode lasers with a homogeneously broadened gain line has recently been the subject of active research. One reason for this research is that such lasers are interesting models of multidimensional dynamic systems with inflexible integral parameters (the total output power, the spectral width, and the gain) and weak internal couplings. The nonlinear dynamics of such a system can be used to study various regimes of motion, such as regular self-excited oscillations, dynamic chaos, and stochastic regimes. Another motivation for this research is that it is of importance to the practical use of these lasers in in-resonator spectroscopy, in generating picosecond and femtosecond pulses, etc.

The dynamics of the emission in the individual modes is determined by the overall gain and loss in the modes, the quantum noise, and the nonlinear interactions of the modes.[1]

For the field amplitude b_q in mode q we can write

$$\frac{db_q}{dt} = -\frac{1}{2}(\gamma + k_q c)b_q + \frac{1}{2}\beta_q b_q + \Phi_q + F_q(t). \tag{1}$$

Here b_q is the dimensionless amplitude in mode q, normalized to satisfy $|b_q|^2 = M_q$, where the right side is the number of photons in the mode. The first term describes the loss, at a rate γ, for all modes. It also describes the in-resonator absorption k_q in the given mode (the absorption in which we are interested here). The second term determines the gain in the active medium with a saturated gain β_q. Since the width (Γ) of the gain band is much greater than the mode spacing $\Delta(\Gamma/\Delta > 10^4)$, we can use a parabolic approximation near the center of the gain band:

$$\beta_q = \frac{\eta\gamma}{1+\frac{J}{J_s}}\left[1-\frac{\Delta^2 q^2}{\Gamma^2}\right], \tag{2}$$

where η is the extent to which the pump exceeds the threshold, $J = \Sigma M_q$ is the total lasing intensity, J_s is the intensity which saturates the working transition, and q is reckoned from the center of the gain band.

The term $\Phi_q(b_1 ... b_q)$ in (1) describes the nonlinear interaction between modes, and F_q is a random force. If F_q is caused by the quantum nature of the light, its normalization is $\langle F_q(t)\, F_{q'}(t')\rangle = \gamma\delta_{qq'}\delta_{tt'}$. When the lasing begins, the spectrum is determined by the spontaneous emission, and the total intensity is reached over a time $1/\gamma \sim 10^{-7}$ s. This intensity is determined by the gain saturation. For the intensities of the individual modes, on the other hand, there is no stabilizing factor such as saturation. The intensity in a mode undergoes random fluctuations, with a typical correlation time $t = \mathrm{M}q/\gamma$, under the influence of a random force. It is this correlation time that determines the maximum effective number of transits in the material under study, i.e., the sensitivity of in-resonator laser spectroscopy.[1,2] This number of transits should increase with an increase in the extent to which the pump exceeds the threshold; at $M \sim 10^7$ it should be ~ 1 s. An absorption coefficient $K = 1/c\tau = 10^{-11}$ cm^{-1} might be detectable in this case.

However, experimental studies have shown that the sensitivity of in-resonator laser spectroscopy using cw lasers falls off when the pump is raised more than a few percent above the threshold.[3] It has also been shown[1] that the lasing switches from a stochastic regime to a dynamic chaos, determined by a nonlinear interaction among modes, as the pump is raised above the threshold. A numerical analysis of Eqs. (1) with (Mandel'shtam–) Brillouin scattering as the nonlinear interaction has shown a good agreement with experimental results. This interaction introduces chaos in the lasing kinetics. It reduces the duration of the fluctuations and thus degrades the sensitivity of in-resonator laser spectroscopy.

To eliminate the effect of Brillouin scattering, we constructed a unidirectional travelling-wave ring laser. The resonator of this cw ring laser was formed by two spherical mirrors and two plane mirrors. The backs of the latter were misaligned 10° to prevent selective effects. Unidirectional lasing was achieved with the help of a nonreciprocal element of MOS-13 glass, with Brewster-angle faces 1.9 cm long. This element was positioned inside a permanent magnet with a field ~1 kG at the axis. In a laser of this type, the dynamic chaos gives way to a regime of regular self-excited oscillations in the intensities of the individual modes. Frames a and b in Fig. 1 show streak photographs of the output spectra of this unidirectional ring laser. The intensity in each mode is seen to oscillate with a period on the order of a millisecond; the actual value of the period depends on the extent to which the pump exceeds the threshold. The phase of the intensity modulation in each mode shifts in the long-wave direction as time elapses, and the overall lasing intensity remains constant. The sensitivity of in-resonator spectroscopy is determined in this case by the typical period of the self-

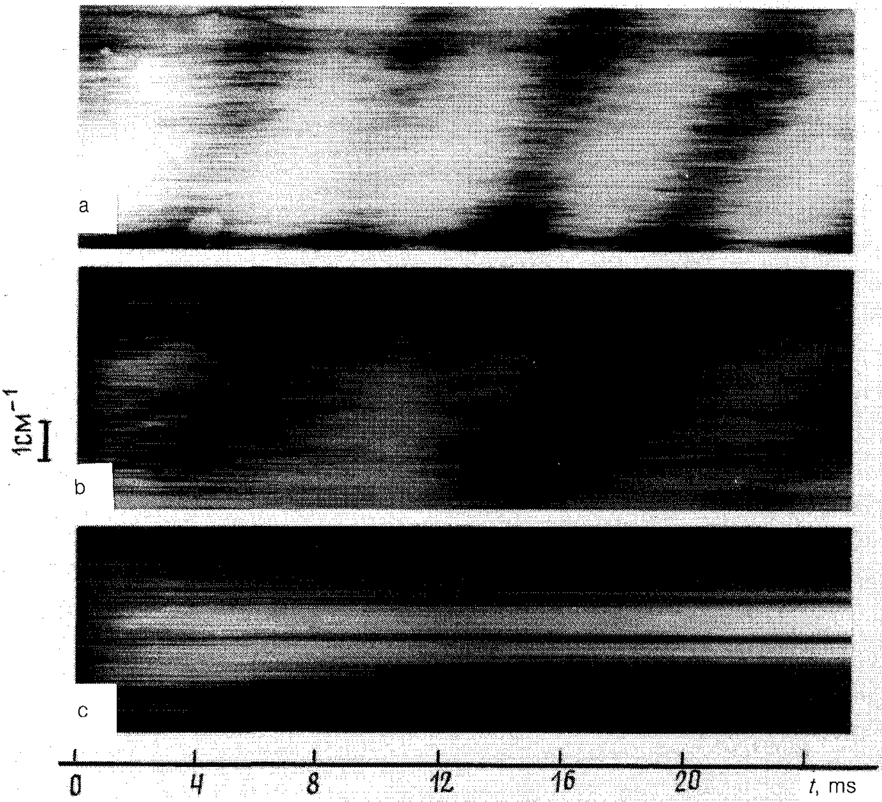

FIG. 1. Streak photographs of the output spectrum of a wide-band ring laser. a,b—The dispersion has not been cancelled ($\delta < 0$), and the extent to which the pump exceeds the threshold is $\eta = 2.1$ (a) or 1.5 (b); c—the dispersion has been cancelled ($\delta = 0$), and $\eta = 1.6$.

excited oscillations. (When the magnetic field at the nonreciprocal element—the Faraday cell—is turned off, chaos arises in the kinetics, and the spectral band of the lasing broadens.)

Analysis of other mechanisms for nonlinear mode interactions leads to the suggestion that a parametric interaction of several unequally spaced modes in the active medium might be the cause of such slow self-excited oscillations in the intensity. The period of such oscillations should be proportional to the parameter δ, which is a measure of the deviation from a uniform mode spacing:

$$\delta = -\left(\frac{\lambda}{L}\right)^3 2\pi c \frac{d^2}{d\lambda^2}\left(\sum n_i l_i\right), \tag{3}$$

where L is the length of the resonator, $\Sigma n_i l_i$ is the sum of the optical lengths of the resonator elements, and λ is the output wavelength. Estimates of the dispersion of the

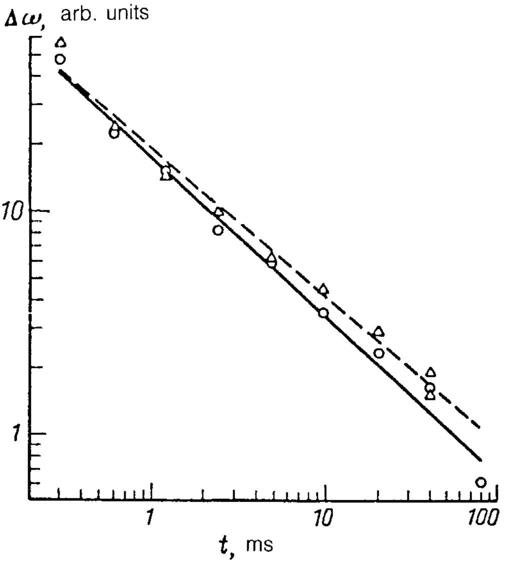

FIG. 2. Spectral width of the output of a laser with cancelled dispersion. Solid line—$\eta = 2$; dashed line—$\eta = 1.2$.

resonator elements (the dye jet, the Faraday-shifter glass, etc.) yield values on the order of 10^{-2} s^{-1} for δ. In the case of equally spaced modes, with a spacing $K\Delta$, the parametric interaction between modes should decay over a time[4] $\sim K\Delta/\gamma$.

To test the hypothesis regarding the effect of a parametric interaction, with allowance for an unequal mode spacing, we introduced a dispersion-cancellation system in the resonator. This was an array of four prisms, similar to one used in the femtosecond range.[5] When the prisms were arranged in positions which corresponded to a cancellation of the dispersion introduced by the term in (3), and which rendered

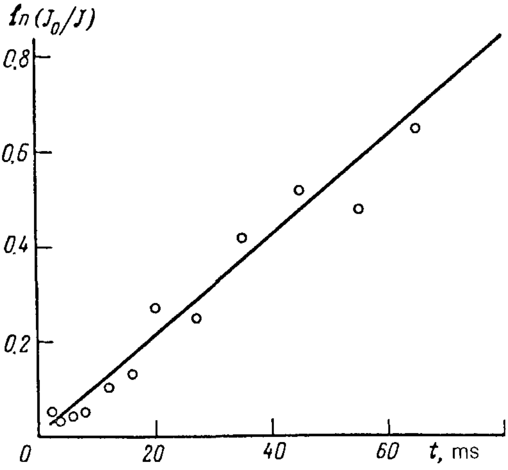

FIG. 3. Time evolution of the depth of the dip in the absorption line for a laser with cancelled dispersion.

the mode spacing uniform, the lasing regime was stabilized (Fig. 1). It can be seen from this figure that the self-excited oscillations in the intensity die out, the output spectrum becomes narrower as time elapses, and the dips corresponding to absorption lines of atmospheric air in the resonator increase in size throughout the lasing pulse. The reason for the narrowing of the spectrum is the parabolic behavior of the gain in (2); this narrowing proceeds in proportion to $t^{-1/2}$. Figure 2 shows the spectral width of the output versus the time. We see that the narrowing of the spectrum continues up to 80 ms. A photodensitometer study of streak photographs of the output spectrum showed that the depth of a dip in a line also increases exponentially with the time, up to at least 60 ms. Figure 3 shows the depth of the dip in the output spectrum for an absorption line with $k = 4\times 10^{-10}$ cm^{-1}, found through a photodensitometer study of a streak photograph of the spectrum. We see that the depth of the dip is 50% at $t = 60$ ms.

We thus see that by suppressing the nonlinear mode interactions we have been able to stabilize the lasing in the individual modes and to raise the sensitivity of in-resonator laser spectroscopy by at least an order of magnitude. In other words, a stabilized laser of this sort makes it possible to measure absorption lines with an absorption coefficient of 10^{-10} cm^{-1} and to detect lines with a coefficient of 10^{-11} cm^{-1}.

[1] Yu. M. Aivazjan, V. V. Ivanov, S. A. Kovalenko *et al.*, Appl. Phys. B **46**, 175 (1988).
[2] V. M. Baev, T. P. Belikova, É. A. Sviridenkov, and A. F. Suchkov, Zh. Eksp. Teor. Fiz. **74**, 43 (1978) [Sov. Phys. JETP **74**, 21 (1978)].
[3] S. J. Harris, J. Chem. Phys. **71**, 4001 (1979).
[4] S. A. Kovalenko, S. P. Semin, and D. D. Toptygin, Kvant. Elektron. (Moscow) **18**, 451 (1991) [Sov. J. Quantum Electron. **21**, 407 (1991)].
[5] R. L. Fork, O. E. Martinez, and J. P. Gordon, Opt. Lett. **9**, 150 (1984).

Translated by D. Parsons