Macroscopic Quantum Fluctuations and First-Order Phase Transition in a Laser

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A homogeneously broadened two-mode ring laser has two metastable states in which one or the other mode intensity is zero. Quantum fluctuations cause the system to switch spontaneously, and at random times, between these states. The probability distribution of the light intensity of one laser mode has been measured, and found to exhibit two completely resolved peaks at zero and nonzero intensities, as predicted. This confirms the existence of a first-order laser phase transition.

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Numerous experiments with single-frequency ring lasers have demonstrated that, when the gain medium is homogeneously broadened, one of the two traveling-wave modes tends to suppress the other one.[1-5] This phenomenon, which appears to have been first predicted by White,[6] is the result of mode competition for the emitted photons. The equations of motion of the two-mode laser exhibit two metastable solutions, in which one or the other mode amplitude is zero. However, in general there is no stable state, and sooner or later a large quantum fluctuation causes the system to switch spontaneously from one metastable state to the other. The mode switching is associated with a discontinuity in the order parameter for the phase transition of the laser field,[7] and therefore corresponds to one of the few cases in which a laser exhibits a first-order phase transition.[4] As a result of the metabistability, the probability distribution $\mathcal{P}(I)$ of the light intensity I of either mode exhibits two peaks, for which evidence has recently been obtained by photon-counting experiments.[4] However, it was not possible to extract the form of $\mathcal{P}(I)$ explicitly from the counting measurements. We now wish to report the results of direct measurements of the probability distribution $\mathcal{P}(I)$, that confirm the existence of the first-order laser phase transition in a quantitative manner.

The equations of motion for the slowly varying complex mode amplitudes $E_1(t)$, $E_2(t)$ of a two-mode laser were already derived by Lamb.[8,9] With the addition of Langevin noise terms $q_1(t)$, $q_2(t)$ to represent the spontaneous emission fluctuations, and in dimensionless units, they take the form

$$\begin{aligned} dE_1/dt &= (a_1 - |E_1|^2 - \xi|E_2|^2)E_1 + q_1 , \\ dE_2/dt &= (a_2 - |E_2|^2 - \xi|E_1|^2)E_2 + q_2 . \end{aligned} \tag{1}$$

a_1, a_2 are dimensionless pump parameters characterizing the excitations, and ξ is the mode coupling constant, which has the value 2 for a homogeneously broadened medium.[9,10] Corresponding to Eq. (1), the joint probability distribution $p(E_1, E_2, t)$ of the two mode amplitudes E_1, E_2 obeys a four-dimensional Fokker-Planck equation, whose steady-state solution can be written[11-13]

$$\begin{aligned} p_s(E_1, E_2) &= \text{const} \times e^{-U} , \\ U &\equiv -\tfrac{1}{2}a_1 I_1 - \tfrac{1}{2}a_2 I_2 + \tfrac{1}{4}I_1^2 + \tfrac{1}{4}I_2^2 + \tfrac{1}{2}\xi I_1 I_2 , \end{aligned} \tag{2}$$

when expressed in terms of intensities $I_1 = |E_1|^2$, $I_2 = |E_2|^2$. The form of the "potential" $U(I_1, I_2)$ is illustrated in Fig. 1 for two different values of ξ. It has a single minimum, corresponding to a stable state with nonzero I_1, I_2 when $\xi = \frac{1}{2}$, but two minima separated by a saddle when $\xi = 2$. Both these minima correspond to highly probable states in which one mode intensity is zero while the other one is nonzero. Quantum fluctuations drive the representative point in phase space from one minimum to the other at random times, and at time intervals that get progressively longer as the pump parameters increase, because the saddle point moves up.[4,5] The light intensity of each mode therefore tends to jump randomly between zero and nonzero values.

By integrating Eq. (2) over one variable, I_2, say, we obtain for the probability distribution $\mathcal{P}(I_1)$ of the other one

$$\mathcal{P}(I_1) = \text{const} \times \exp[\tfrac{1}{4}(\xi^2 - 1)I_1^2 - \tfrac{1}{2}(a_2\xi - a_1)I_1 + \tfrac{1}{4}a_2^2][1 - \exp(\tfrac{1}{2}\xi I_1 - \tfrac{1}{2}a_2)] . \tag{3}$$

The form of $\mathcal{P}(I_1)$ is illustrated in Fig. 2(a) for $\xi = 2$, $a = 9$, $\Delta a = 0.29$. It has two branches, corresponding to one peak at $I_1 = 0$, the low-intensity peak L, and a high-intensity peak H at $I_1 = I_H$, with

$$I_H \approx a - 4/(a + \tfrac{3}{2}\Delta a) , \tag{4}$$

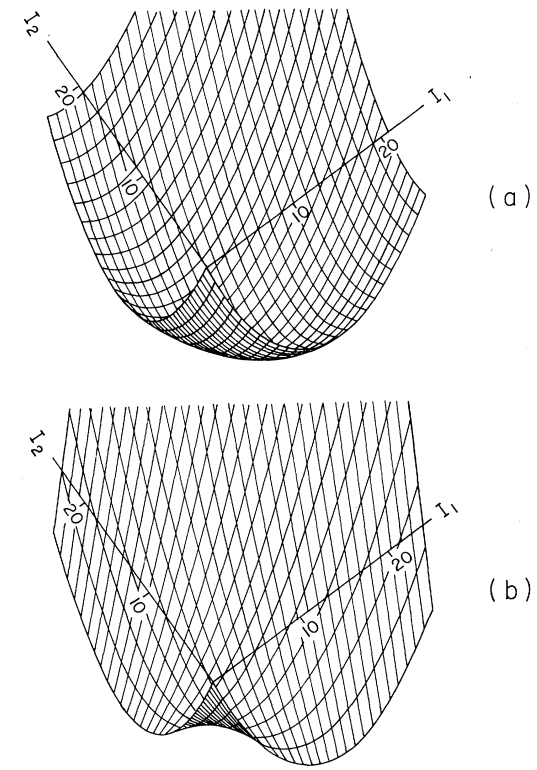

FIG. 1. The form of the potential $U(I_1, I_2)$ with $a_1 = 15$, $a_2 = 14.5$ for coupling constants (a) $\xi = \frac{1}{2}$; (b) $\xi = 2$.

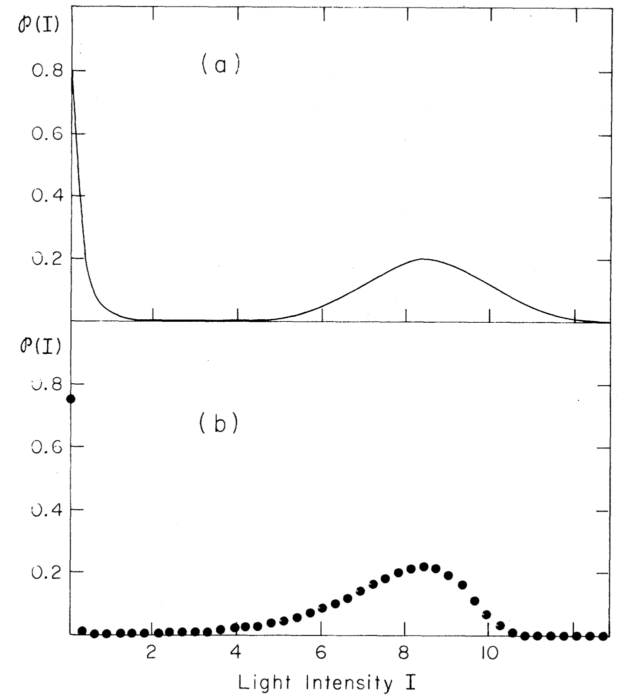

FIG. 2. The form of the probability distribution $\mathcal{P}(I)$ with $\xi = 2$, $a = 9$, $\Delta a = 0.29$. (a) Computed from Eq. (3); (b) measured.

where $a \equiv \frac{1}{2}(a_1 + a_2)$, $\Delta a \equiv a_1 - a_2$. The areas under these two peaks yield the probabilities P_L or P_H that the light intensity I_1 is low or high, respectively, and it can be shown from Eq. (3) that they are given by[4]

$$\left.\begin{array}{l} P_L \\ \\ P_H \end{array}\right\} \simeq [1 + e^{\pm \Delta a/2}(a \mp \tfrac{3}{2}\Delta a)/(a \pm \tfrac{3}{2}\Delta a)]^{-1} \qquad (5)$$

while their ratio is

$$P_H/P_L \approx e^{a\Delta a/2 - 3\Delta a/a}. \qquad (6)$$

For a symmetric two-mode laser with $\Delta a = 0$, $P_L = \frac{1}{2} = P_H$. Also from Eq. (3) we have the ratio

$$\mathcal{P}(I_H)/P_H \approx 0.28, \qquad (7)$$

whereas the ratio of the L to H peak heights is approximately given by

$$\mathcal{P}(0)/\mathcal{P}(I_H) \approx \pi^{1/2} a \exp(-\tfrac{1}{2}a\Delta a + \tfrac{3}{2}\Delta a/a - 4/a^2). \qquad (8)$$

As a increases, this decreases from values greater than unity to values below unity when $\Delta a > 0$. The most probable value of the light intensity I_1, which is an order parameter for the phase transition, therefore changes discontinuously from zero to nonzero values.

We have measured the probability distribution $\mathcal{P}(I)$ directly for a dye ring laser, by aiming the laser beam on a photodiode and sampling the output of the diode at regular intervals with the aid of a pulse-height analyzer. Figure 3 shows an outline of the apparatus. The active laser medium is rhodamine-6G dye in methanol and water,

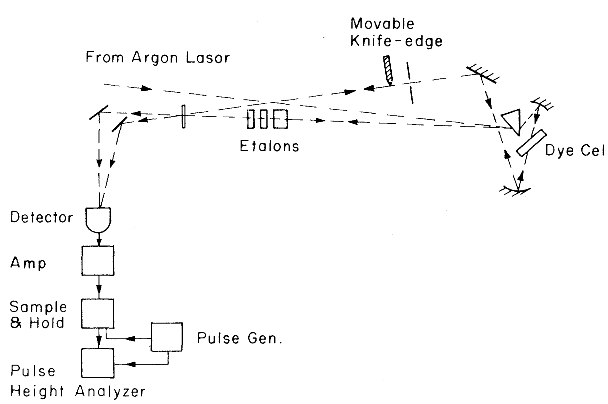

FIG. 3. Outline of the apparatus.

that is circulated at high velocity through a cell, and is optically pumped by an argon-ion laser. A movable knife edge acts as a variable loss, that allows the working point of the laser to be controlled. The three etalons shown ensure single-frequency, two-mode operation. Two light beams emerge in slightly different directions from the output mirror of the ring laser, corresponding to the two counterpropagating modes, and one or the other is directed to the photodiode. The amplified output of the diode is proportional to $I_1(t)$ or $I_2(t)$ to a good approximation. The fluctuating detector signal is passed to a sample-and-hold circuit, that is activated by an external sampling pulse at regular intervals of 1 msec, and then maintains its output level long enough for it to be channeled to the appropriate bin of a multichannel pulse-height analyzer. Each channel n ($n=0,1,2,\ldots,100$) corresponds to a certain light intensity I, and the number of events N_n accumulated in channel n eventually provides a measure of the probability density $\mathcal{P}(I)$ through the relation

$$\mathcal{P}(I)\delta I = N_n / \sum_n N_n , \tag{9}$$

where δI is the channel width. Typical values of N_n range from 10^4 or 10^5 near the peaks to less than 10 between peaks.

In order to relate the experimental results to the theoretical Eq. (3), we need to determine the two pump parameters a_1 and a_2 or a and Δa, and also the channel width δI. We first calculate the probabilities P_H and P_L by summing the counts N_n falling under each peak separately, and dividing by the total number $\sum N_n$. We then use Eqs. (7) and (9) to determine δI. Equation (4), which is rather insensitive to the value of Δa, then yields a, whereupon Eq. (6) can be solved for Δa. This procedure avoids use of the value $\mathcal{P}(0)$ determined from the experiment, which is most uncertain, as we point out below.

We might point out that a dye laser is sometimes turned on and off as a result of completely spurious causes, such as bubbles within the dye solution. Such spurious effects may play a role far above threshold, when the spontaneous switching periods become seconds or minutes long, but they are usually unimportant in the threshold region, where the switching times are of order 10 msec. Moreover, bubbles are unlikely to result in complete anticorrelation between the two modes,[4] or in the very strong dependence of the switching period on pump parameter, such as we have observed,[5] and as predicted by the equations of motion (1).

In Fig. 2 some experimental values of $\mathcal{P}(I)$ are compared with the theoretical probability distribution given by Eq. (3) for $\xi=2$, $a=9$, $\Delta a=0.29$, $\delta I=0.30$. The two distributions are qualitatively similar, although there are quantitative differences. In practice the high-intensity tail of each peak falls to zero more rapidly than the theory predicts. Indeed, the peak at $I=0$ decays almost to zero in less than one analyzer channel width δI, so that $\mathcal{P}(0)$ given by Eq. (9) is not really the differential probability density, but an integral over δI. We believe that the suppression of small values of the light intensity is probably attributable to backscattering from one ring laser mode into the other, for which evidence has already been encounted.[4] It is also conceivable that the existence of triplet states in the dye molecule plays some role in causing departures from the usual laser theory.[14] However, the measured probability distribution clearly exhibits the predicted two, highly probable, metastable laser states, with a region of almost zero probability in between. As the pump parameter is raised, $\mathcal{P}(I_H)$ is found to increase gradually while $\mathcal{P}(0)$ falls. Eventually $\mathcal{P}(I_H)>\mathcal{P}(0)$, at which point the most probable light intensity jumps from 0 to a nonzero value, and this implies the existence of a discontinuity of the order parameter for the phase transition.

Spontaneous on-off switching of the two mode intensities is one of relatively few direct macroscopic manifestations of quantum fluctuations in physics. A single, spontaneously emitted photon can cause two rather intense laser beams to be turned on and off in this experiment.

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Oscillatory Phenomena and *Q* Switching in a Model for a Laser with a Saturable Absorber

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Sufficiently long population decay times and sufficiently short dipole decay times in a single-mode laser with saturable absorber permit passive Q switching in the form of a hard-mode sustained relaxation oscillation.

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A saturable absorber in a cavity permits emission of trains of pulses (passive Q switching).[1,2] We show that this phenomenon appears as a limit-cycle oscillation in a model of a laser with absorber.[3,4] Using the notation introduced in Ref. 3 we have

$$\dot{E} = Nv + \overline{N}\overline{v} - \kappa E, \tag{1a}$$

$$\dot{v} = |g|^2 DE - \gamma_\perp v, \tag{1b}$$

$$\dot{\overline{v}} = |g|^2 \overline{D}E - \overline{\gamma}_\perp \overline{v}, \tag{1c}$$

$$\dot{D} = -\gamma_\parallel D - 4vE + \gamma_\parallel \sigma, \tag{1d}$$

$$\dot{\overline{D}} = -\overline{\gamma}_\parallel \overline{D} - 4\overline{v}E + \overline{\gamma}_\parallel \overline{\sigma}. \tag{1e}$$

A dot over a quantity indicates time derivative. E is the electric field; D is the atomic inversion; v is the polarization; $\gamma_\parallel$ and $\gamma_\perp$ are longitudinal and transverse relaxation rates, respectively; κ is the photon decay rate in the cavity, $\kappa = c(1-R)/L$, where R is the reflectivity of the mirror at the boundaries and L is the length of the cavity; σ is the unsaturated inversion; N is the number of atoms; and $|g|$ is the field-matter coupling constant. A bar over a quantity refers to the passive atoms. c is the velocity of light.

We now write

$$E = -(\gamma_\parallel \gamma_\perp)^{1/2} a / 2|g|,$$

$$v = -\sigma |g| (\gamma_\parallel / 4\gamma_\perp)^{1/2} p, \quad \overline{v} = -\overline{\sigma}|g| (\gamma_\parallel/4\gamma_\perp)^{1/2}\overline{p},$$

$$D = \sigma(1-d), \quad \overline{D} = \overline{\sigma}(1-\overline{d}), \quad t' = \gamma_\perp t, \quad \omega = \gamma_\parallel/\gamma_\perp,$$

$$r_1 = \overline{\gamma}_\perp/\gamma_\perp, \quad r_2 = \overline{\gamma}_\parallel/\gamma_\parallel, \quad \rho = \kappa/\gamma_\perp,$$

$$A = N|g|^2\sigma/\kappa\gamma_\perp,$$

and

$$C = 1 - \overline{N}|g|^2\overline{\sigma}/\kappa\gamma_\perp.$$

We obtain the following dimensionless equations:

$$\dot{a} = \rho[-a + Ap + r_1(1-C)\overline{p}] \tag{2a}$$

$$\dot{p} = a(1-d) - p \tag{2b}$$

$$\dot{\overline{p}} = a(1-\overline{d}) - \overline{p}r_1 \tag{2c}$$

$$\dot{d} = \omega(-d + ap) \tag{2d}$$

$$\dot{\overline{d}} = \omega(-r_2\overline{d} + a\overline{p}). \tag{2e}$$

We set $r_2 = 1$, i.e., we take the longitudinal relaxation time, $T_1 = 1/\gamma_\parallel$, the same for both active and passive atoms which is consistent with the assumption of resonance between the emitting and absorbing transitions. We fix $\omega = 0.01$, i.e., we take the transverse relaxation time, $T_2 = 1/\gamma_\perp$, to be two orders of magnitude smaller than T_1. We also take $\rho \simeq 0.1$ and $\omega < \rho$. For related problems with, however, different parameter ranges see Knapp, Risken, and Wollmer.[5]

The pumping rates are hidden in A and C. Without absorber the threshold for laser behavior is at $A = 1$, and the steady-state laser intensity grows with $A - 1$. The absorbing medium is pumped as an emitter if C is smaller than unity, with threshold at $C = 1 - 1/r_1$. Thus we restrict C to be larger than unity.

For

$$(1-r_1)^{-1} \equiv C_{tc} \leq C \leq (\rho + r_1)/\rho(1-r_1) \equiv C_{osc},$$

the zero-field solution is stable up to $A = C$ where