Self-Pulsing in Intrinsic Optical Bistability with Two-Level Molecules

Bernard Segard and Bruno Macke
Laboratoire de Spectroscopie Hertzienne de l'Université de Lille, Unité Fondamentale de Recherche de Physique,
59655 Villeneuve d'Ascq Cédex, France

(Received 10 August 1987)

Well-developed sine-wave self-pulsing has been observed in the beam transmitted by a passive cavity containing a molecular gas, subjected to a cw incident beam. The free spectral range of the cavity is comparable to the Rabi frequency inside the cavity and much larger than the relaxation rate. The self-pulsing is attributed to the multimode instability predicted by Bonifacio and Lugiato in 1978 but not yet observed.

PACS numbers: 42.65.−k, 33.80.−b, 42.50.−p

Optical bistability[1,2] is a remarkable example of cooperative behavior in an open system far from thermal equilibrium. From this viewpoint, the search for instabilities in optical bistable devices is a crucial point. Bistability itself originates in the fact that the intermediate branch of the S-shaped bistability curve is unstable, but it was early recognized[3] that the positive-slope regions of this curve may also be unstable for suitable values of the control parameters, leading to a much richer phenomenology, including regenerative oscillations,[3] self-pulsing,[4] and chaos.[5]

Among the different types of optical bistable devices, the cavity filled with two-level atoms (or molecules) and driven by a cw incident beam appears to be of special importance.[2] It is a canonical model in nonlinear optics and, if necessary, is liable to a fully quantum treatment. Following the pioneering works by Bonifacio and Lugiato[4] and by Ikeda,[5] numerous types of instabilities have been predicted to occur in this system[6] but only one of them has been actually observed. This has been achieved in an experiment in which two-level atoms couple to a single mode of the cavity (single-mode instability).[7] The instability originates then in a subtle interplay of nonlinear gain and dispersion in the medium in order that multiple frequencies can coexist in the cavity with effectively only a single mode.[7] In contrast, multimode instability, which has been the first to be predicted[4,5] and which may be more simply understood in terms of side-mode gain,[8,9] has not yet been observed with continuous-wave excitation.[10] We succeed in observing instability in such conditions, achieving an experiment in which the free spectral range of the cavity is comparable to the Rabi frequency (power broadening) inside the cavity and much larger than the inverse of the medium response time. The instabilities observed in this case are then expected to be closely related to the self-pulsing initially predicted by Bonifacio and Lugiato[4] and studied extensively in subsequent papers.[2,11,12] As noted by Gibbs,[13] this self-pulsing seems to be the signature of Rabi flopping, resulting in side-mode gain.

Our bistable system[14] is a Fabry-Perot cavity of length $l=182$ m (free spectral range $c/2l=830$ kHz) filled with $HC^{15}N$ at low pressure (0.5-1.5 mTorr). The cavity is cw driven by a source phase locked at a frequency ν_s close to the frequency ν_m of the $J=0$-1 rotational line of $HC^{15}N$ ($\nu_m=86.05496$ GHz, wavelength $\lambda\cong3.5$ mm). Between the input and output mirrors of reflectivity $R_0=0.95$ and transmittivity $T_0=0.05$ (negligible losses), the beam is guided (except for short sections) by a very oversized helix waveguide (helix radius $a=30$ mm). This waveguide acts as a mode filter and transmits only the modes TE_{0n} of electric field $E_r=E_z=0$ and $E_\theta=E_0J_1(rj_{1n}/a)$, where J_1 is the first-order Bessell function and j_{1n} its nth zero. The input and the output couplings are made in the single-mode TE_{01}, presenting the lowest losses. The power transmission in this mode is $A_0=0.73$ per 182-m trip. It is easily shown that our cavity is equivalent to a cavity without distributed losses provided that R_0 and T_0 are replaced by $T_e=T_0\sqrt{A_0}=0.043$ and $R_e=R_0A_0=0.69$. This leads to a mode width of 48 kHz (HWHM) and to a finesse $F=8.5$.

The 0-1 rotational line of $HC^{15}N$ is easily saturated at moderate power level on account of the large permanent dipole of $HC^{15}N$ ($\mu\cong3$ D) and, if we neglect its narrow magnetic hyperfine structure (16 kHz), is characterized by a unique Rabi frequency[15] $\nu_R=\mu_{01}E/h=\mu E/h\sqrt{3}$, that is, about 750 kHz for the mean power density of 1 mW/cm^2 typically achieved inside the cavity in our experiments. This value being significantly larger than the inhomogeneous Doppler broadening ($\cong100$ kHz), the $HC^{15}N$ gas approximates closely an ideal medium of homogeneously broadened two-level systems. The relaxation is mainly collisional and, as usual for rotational relaxation, is characterized by a unique rate γ ($\gamma_\perp=\gamma_\parallel$), proportional to the gas pressure ($\gamma/2\pi\cong23$ kHz at 1 mTorr). Finally, the large power absorption of the line in the linear regime ($\alpha_\infty\cong0.8$ m^{-1} in the collisional limit[16]) leads to a cooperativity parameter $C=\alpha_\infty lF/2\pi\cong200$, that is much beyond the threshold of bistability.

Figure 1 gives an example of bistability cycle, evidencing a well-developed self-pulsing on its upper branch. It

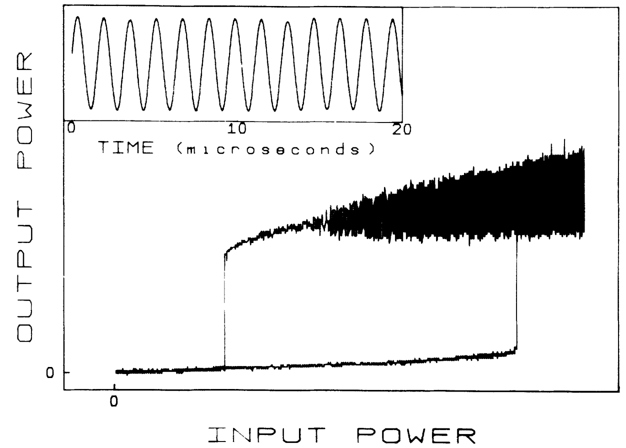

FIG. 1. Bistability cycle observed for $\nu_c = \nu_m$, $\nu_s - \nu_m = 317$ kHz. Gas pressure $p \simeq 0.75$ mTorr. Inset: Sine-wave self-pulsing obtained when the incident power is fixed at its maximum.

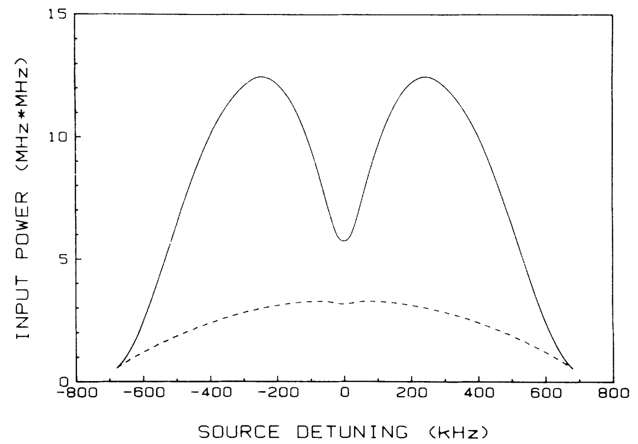

FIG. 2. Upper (full line) and lower (dotted line) critical switching powers vs $\nu_s - \nu_m$, computed for $\nu_c = \nu_m$ and $p \cong 1.3$ mTorr. The power is given by the square of the corresponding Rabi frequency ν_R^I.

has been obtained by our adiabatically sweeping the input power, controlled by a *p-i-n* diode modulator. The molecular frequency ν_m coincides with that of a longitudinal mode of the cavity ν_c but differs from that of the source. For a fixed input power (cw excitation), the self-pulsing is purely sinusoidal—this point has been verified ***in all our experiments*** by a spectral analysis—and its frequency is nearly constant inside the instability domain, increasing up to 630 kHz for the maximum available incident power (estimated at 50 mW just before the input mirror).

Except obviously for the self-pulsing, the hysteresis cycle of Fig. 1 is fairly well fitted by the steady-state bistability curve of a ring cavity (with $\mathcal{L} = L = 364$ m in the notations of Ref. 2, $R = R_e$, $T = T_e$), computed in a plane-wave approximation without any adjustment of the experimental parameters. This surprising agreement has been confirmed on hysteresis cycles obtained in different conditions.[17] The ratio of the upper and lower switching powers in Fig. 1 is exactly reproduced by the adjustment of only the gas pressure p (0.6 mTorr instead of 0.75 mTorr). For the maximum incident power, the Rabi frequencies, derived from the calculation, are $\nu_R^I = 1.33$ MHz and $\nu_R^c = 0.45$ MHz just before the input and output mirrors, respectively. ν_R^I is quite consistent with the estimated available power (50 mW) and ν_R^c gives an underestimate of the rms Rabi frequency ν_R inside the cavity.

In all our experiments, self-pulsing has been only observed in the presence of bistability, on the upper branch of the bistability curve. By our sweeping the input power (standard procedure), this branch is attained only when the available input power P_I is larger than the *upper* switching power P_U. A much wider domain can be explored by use of the source frequency ν_s as control parameter.[18] Starting from a source detuning such that the upper and lower critical powers (P_U and P_L) coalesce (no bistability) at a very low level (Fig. 2), it is indeed possible to follow by continuity the upper branch as long as P_I is larger than P_L which may be 1 order of magnitude lower than P_U ($C \gg 1$). Moreover, by removal of the *p-i-n* modulator (insertion loss $\simeq 3$ dB), no longer required, the input Rabi frequency can be significantly enhanced ($\nu_R^I \cong 1.9$ MHz).

Figure 3 has been obtained in these conditions for $\nu_m = \nu_c$. Figure 3(a), given for reference, shows three successive TE_{01} modes of the empty cavity. Note the absence of parasitic modes. With introduction of the $\mathrm{HC}^{15}\mathrm{N}$ gas, self-pulsing is observed for 150 kHz $< |\nu_s - \nu_m| < 370$ kHz. If the source frequency ν_s is fixed (cw excitation), the self-pulsing is purely sinusoidal as already mentioned and its frequency is nearly constant in the instability domain ($\cong 670$ kHz). The four recordings of Fig. 3 being obtained with the same sensitivity, the Rabi frequency inside the cavity ν_R^c can be easily derived in any point of the curves *a*, *b*, *c*, and *d* from its value on resonance without gas $[= \nu_R^I \sqrt{T_e}/(1 - R_e) \cong 1.3 \text{ MHz}]$. ν_R^c is nearly constant in the unstable regions of *b*, *c*, and *d* ($\nu_R^c \cong 850$ kHz). Note the weak gas absorption in these regions ($\langle \alpha l \rangle \cong 0.15$), as expected because of the strong saturation. Moreover let us remark that the attenuation and the shift of the sidemodes in Figs. 3(b), 3(c), and 3(d) are quite consistent with the estimated Rabi frequencies.

Self-pulsing is generally detected with the best signal-to-noise ratio in "resonant configurations" ($\nu_m = \nu_c$, Figs. 1 and 3), with the whole available power and pressures ranging from 1 to 1.5 mTorr. We have, however, explored quite different situations. For instance, Fig. 4 shows self-pulsing (frequency $\cong 400$ kHz, $\nu_R^c \cong 340$

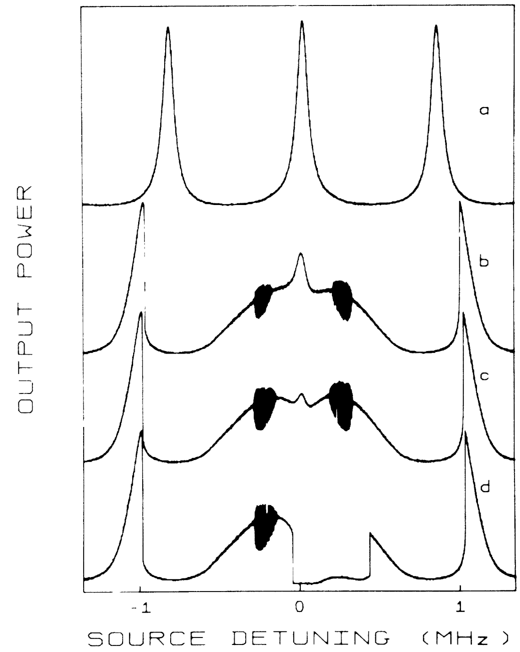

FIG. 3. Transmission of the cavity vs $\nu_s - \nu_m$ for $\nu_m = \nu_c$ and $p \cong$ (a) 0, (b) 1.3, (c) 1.35, and (d) 1.4 mTorr. For $p = 1.4$ mTorr, the bistable system switches down at ν_s such that $P_L < P_l$ and switches up back only when $P_l > P_U$ (Ref. 18). The symmetric curve is obtained reversing the frequency sweep (Ref. 17). At still higher pressures, the condition $P_L < P_l$ is no longer fulfilled inside the instability domain and the self-pulsing cannot be observed.

kHz) observed in an "antiresonant configuration" (ν_m halfway between two cavity resonances). The exploitation of numerous recordings obtained in various experimental conditions allows us to specify the following points.

(i) Self-pulsing has been observed for source detunings down to about 100 kHz but never in the case of purely absorptive bistability ($\nu_s = \nu_m$). This is in agreement with theoretical calculations predicting that self-pulsing does not occur in this case because of the transverse distribution of the field[19,20] but remains in the case of mixed absorptive and dispersive bistability.[12] Let us also mention that the power threshold of instability is lowered in detuned configurations according to the theory.

(ii) The self-pulsing frequency ranges from 400 (see, e.g., Fig. 4) to 730 kHz (for $\nu_m - \nu_s = 240$ kHz, $\nu_c - \nu_s = 100$ kHz, $p = 1.2$ mTorr). Contrary to what is expected for single-mode instability,[7] it is not strongly correlated with the cavity detuning $\nu_s - \nu_c$ (ranging from 100 to 500 kHz).

(iii) At low pressures (<1 mTorr), the instabilities may appear only in a restricted range of input power, as predicted theoretically.[12]

To conclude, we have observed sine-wave self-pulsing in intrinsic optical bistability. The free spectral range of the cavity being comparable to the Rabi frequency inside the cavity but large compared to the relaxation rate γ, there is a strong suspicion that this is the first experimental observation of the multimode instability initially predicted by Bonifacio and Lugiato.[4] The difference between the self-pulsing frequency and the free spectral range can be probably explained by departures of our system from canonical conditions (mean-field limit). This point is being checked by numerical integration of the Bloch-Maxwell equations of the "equivalent" ring cavity.

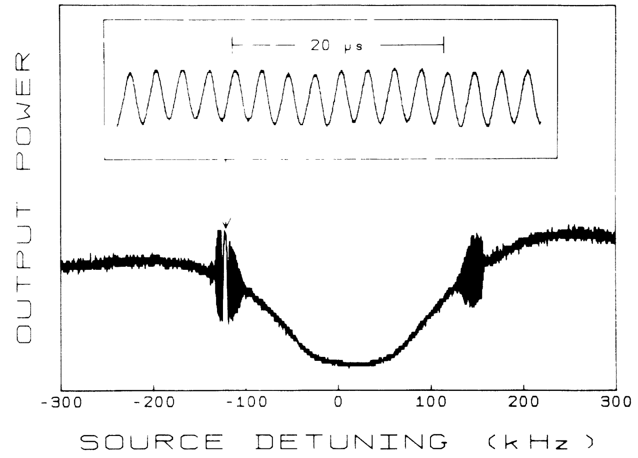

FIG. 4. Observation of self-pulsing in the antiresonant configuration ($\nu_m = \nu_c + c/4l$). Pressure $p = 0.85$ mTorr. Other parameters as in Fig. 3. Inset: Self-pulsing for a fixed source frequency.

The Laboratoire de Spectroscopie Hertzienne is a Unité Associée au Centre National de la Recherche Scientifique (France). This work was supported by the European Economic Community under Contract No. ST2J-0187F (Stimulation Action) and by the Région Nord-Pas-de-Calais. We gratefully acknowledge assistance of J. Zemmouri in the experiments. A preliminary account of this work was given at the Workshop on Instabilities, Dynamics, and Chaos in Nonlinear Optical Systems, 8-10 July 1987, Il Ciocco, Lucca, Italy.

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[15] By numerically solving the Bloch equations, we have verified that the hyperfine structure plays no role provided that its width is small compared to ν_R (see Zemmouri, Ref. 14).

[16] Measured value. Note that, as a result of saturation, the actual absorption coefficient is typically 3 orders of magnitude below α_∞.

[17] In some cases, a second bistability cycle appears both on the computed and observed curves (multistability). The corresponding output levels are very low.

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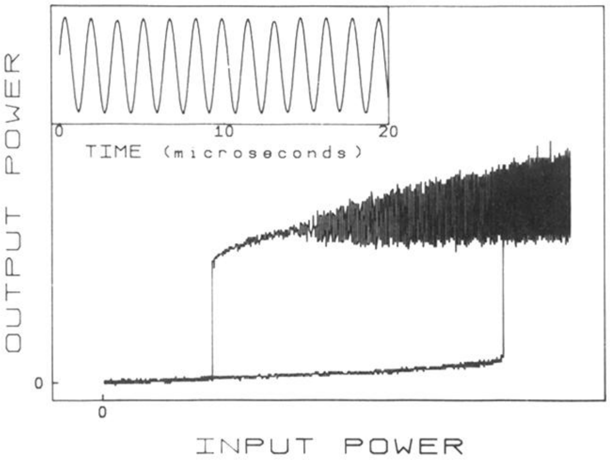

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