ON THE EXPERIMENTAL ACCESSIBILITY OF THE SELF-PULSING REGIME OF THE LORENZ MODEL FOR SINGLE MODE HOMOGENEOUSLY BROADENED LASERS

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The transverse and longitudinal relaxation rates of optically pumped molecular lasers are examined with respect to the self-pulsing solutions predicted by the Lorenz model. We conclude that even in the limit of strong inversion effects (NH_3), the required ratio $\gamma_{\parallel}/\gamma_{\perp} \leqslant 0.2$ is not accessible. This suggests that the experimental measurements of self-pulsing in these lasers are not a definitive observation of a Lorentz-type instability.

1. Introduction

In the recent past a great deal of effort has been put into the experimental verification of the single-mode instability predicted by the mean field Maxwell-Bloch equations [1,2]. Since the elucidation by Haken that the equations for a resonantly tuned laser were isomorphic to the Lorenz equations, new regimes of the control space became accessible when lasers are the object under study [2].

The Lorenz model has been studied in the fluid limit and is known to predict a sudden transition from stable fixed points to chaotic behavior when the system reaches the second laser threshold [3]. This behavior clearly predicts no route to chaos, but a first order type transition for the appearance of a strange attractor.

Recent work has shown that in the limit of low $\tilde{\gamma} = \gamma_{\parallel}/\gamma_{\perp}$, that periodic pulsing solutions to the Lorenz equations exist [4]. These solutions still require that the laser be nine to ten times above threshold and that $\tilde{\gamma} \leqslant 0.20$. It has also been conjectured that this limit of $\tilde{\gamma} < 0.20$ is experimentally accessible and is substantiated by the observed self-pulsing of the unidirectionally pumped $^{15}NH_3$ system [5,6]. Unfortunately, this system is likely to have a $\tilde{\gamma}$ of the order of unity. The experimental results on $^{14}NH_3$ [7] can only be compared to models where detuning is included [8] and are therefore not relevant to the Lorenz instability. Furthermore, the theory in ref. [8] clearly shows sensitivity to control parameter dynamics, resulting in nested thresholds for different approaches to a new control space point. In the following section we discuss the problem of obtaining systems where $\tilde{\gamma} < 0.2$ and show via a simple classical picture that is substantiated by experimental measurements, that this value is not accessible in currently known molecular systems.

2. Molecular relaxation

It is clear from the work in ref. [4] that $\tilde{\gamma}$ is a viable control parameter and is a molecular property, not a laser property. This means that some degree of serious rigor must be put forth justifying a particular $\tilde{\gamma}$ value from molecular data, not laser behavior, if a Lorenz instability of any kind is claimed.

Molecules in the far infrared emit photons in the energy range of 0.5–0.05 kT when room temperature experimental conditions prevail. This small energy is what has led to the large success of the diabatic collision models for line broadening of symmetric tops molecules [9–12]. There are only small differences

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between impact theories and theories which include full resonance interactions. This is primarily due to the large hard sphere extent of molecules and tremendous level dilution in systems with many degrees of freedom. The facts stated above may be substantiated by various measurements on *ground vibrational state*, relaxation rates of $^{15,14}NH_3$ [13–16] and other polyatomic molecules [17]. The conclusions are that these systems have $\tilde{\gamma}$ values of the order of unity.

The exceptions to the above stated behavior are molecules with inversion tunnelling effects. In principle all nonplanar species can exhibit inversion, however, most systems have extremely low inversion frequencies or splittings. The AsH_3 molecule which has the same form as NH_3, tunnels from one As position to the other once every two days in the ground ν_2 vibrational state nad once every day in the first excited ν_2 vibrational state [18]. This extremely slow tunneling is due to the large barrier the H_3 ring presents to the As atom. NH_3 on the other hand has a very low barrier and can tunnel much more readily. The ground ν_2 state has an inversion frequency of about 3×10^{10} Hz while the excited vibrational state has a frequency of about 10^{12} Hz [13].

The consequences of inversion on collisional energy transfer ($\gamma_\parallel$) become important when the molecule inverts faster than the duration of a collision. The effect of inversion on dephasing ($\gamma_\perp$) are not greatly felt since all that is required is phase interruption which does not require dipole alignment throughout the duration of a collision.

All rotational levels of $^{14,15}NH_3$ are inversion doublets *except* those with $K = 0$ quantum numbers. These levels are not doublets as either the symmetric or antisymmetric level alone is allowed by the Pauli exclusion principle. This has great consequence to the $\tilde{\gamma}$ values of the line used in experiments on the self-pulsing $^{15}NH_3$ laser which has been claimed to exhibit the symmetric envelope solution in ref. [4]. Since the laser transition follows $\Delta K = 0$ selection rules both the pumping and lasing processes occur via $K = 0$ levels [19]. This fact implies that $\tilde{\gamma}$ for the lasing transition in $^{15}NH_3$ is of the order of 1.

A simple classical picture which explains the lowering of $\gamma_\parallel$ with inversion can be formulated on the basis of an effective cross-section. Since the excited molecules require a dipole coupled collision with the inverting atom in order to cause a change in rotational energy, the situation where the atom is tunneling from one side to the other of the rotating molecule clearly presents a smaller area for collision. If R_0 is the effective collision cross-section distance of the N atom from the center of the H_3 plane, then a classical approximation to the inverting molecule is given by

$$R(t) = R_0(1 + \cos \Delta t), \qquad (1)$$

where Δ is the inversion frequency. The collisions which are effective in a transfer of rotational energy can be associated with a collision time, T_c, which is of the order of $2R_0/v(\approx 10^{-12}\ \mathrm{s})$ where v is the average molecular velocity. The effective radius of the rotating and inverting NH_3 molecule may be found from

$$\langle R \rangle_{T_c} = \frac{R_0}{T_c} \int_0^{T_c} (1 + \cos \Delta t)\, dt. \qquad (2)$$

This results in a ratio of collisional relaxation rates in the excited and ground vibrational states ($K \neq 0$) given by

$$\frac{\gamma_\parallel^e}{\gamma_\parallel^g} \approx \left(\frac{1 + |\sin \Delta_e T_c|/2\Delta_e T_c + |\sin \Delta_g T_c|/2\Delta_g T_c}{1 + |\sin \Delta_g T_c|/\Delta_g T_c} \right)^2 \qquad (3)$$

Δ_e is the inversion frequency of the excited vibrational state and Δ_g is the inversion frequency of the ground vibrational state. When the $^{14}NH_3$ values are inserted into eq. (3), the value $\gamma_\parallel^e/\gamma_\parallel^g \approx 0.30$ is obtained. This is in good agreement with the $\tilde{\gamma} = 0.29$ value measured for the $^{14}NH_3$ laser levels, assuming that $\gamma_\perp$ is essentially vibrational state independent [13]. This claim is experimentally supported in ref. [13] for all the J, K levels studied.

Another interesting result of eq. (3) is that when $\Delta_e \to \infty$ and $\Delta_g \to 0$, the limiting ratio of 0.25 is obtained. This is expected to be a lower bound for $\tilde{\gamma}$ for rotational transitions in the far infrared. The results of numerical simulation of the Lorenz equations for this case show that a direct transition into chaos is expected. Figs. 1a and 1b show the corresponding time and phase plane behavior for $\tilde{\gamma} = 0.25$ once the second threshold is reached. Furthermore, it should be pointed out that for other systems where inversion is not present, $\gamma_\parallel$ would be expected to increase in the excited vibrational state owing to the increased collision radius due to vibrational well anharmonicity

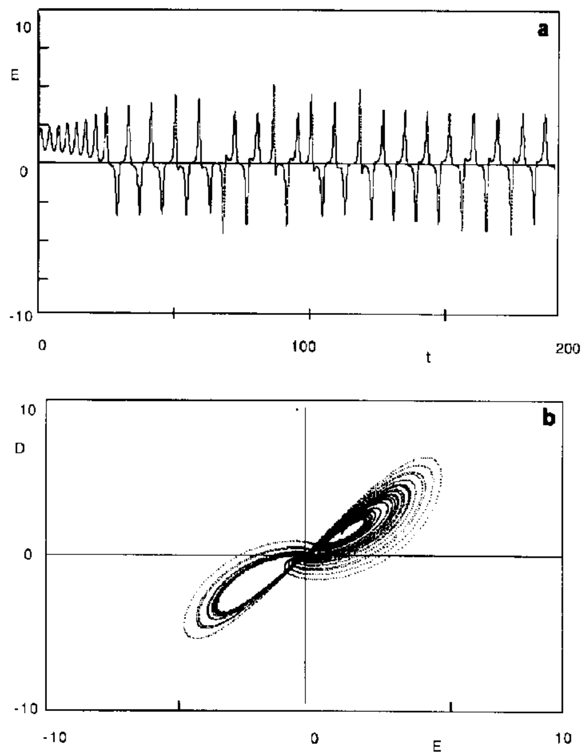

Fig. 1. Chaotic time evolution (a) and phase plane portrait (b) for $\tilde{\gamma} = 0.25$ once the second threshold is reached. Time is in units of $\gamma_\perp^{-1}$.

[20]. This latter effect results in a few percent increase and is responsible for transport effects such as laser induced diffusion [21].

3. Conclusions

We have shown that the $^{15}NH_3$ laser (374 μm) transition must have a γ value near unity based on the Pauli exclusion principle and a didactic model for the effects of inversion on the $\gamma_\parallel$ rate. In addition, we point out that the experimentally known relaxation rates for the $^{14}NH_3$ laser (81 μm) transition result in a $\tilde{\gamma} = 0.28$. Therefore, neither of these lasers is capable of accessing the parameter space required for the self-pulsing regime of the Lorenz equations. Based on these facts, we urge experimentalists and theorists to take caution when claims are made concerning the observation of a Lorenz-type instability. It is know that the experimental far infrared systems used to date have strong spatial variations due to the effects of pump absorption and pump transverse profiles. This situation has resulted due to the experimentalists' attempt to eliminate backward propagating pump waves and inhomogeneous broadening due to ac Stark effects. However, we believe that the effects of ac Stark broadening due to the coherence of the pumping process [22] may be desirable and not a liability. The pump ac Stark effect increases $\gamma_\perp$ only and can result in an effectively lower $\tilde{\gamma}$, allowing for pulsing solutions. In addition, the increased pump intensity may be utilized to increase the uniformity of the gain volume, bringing the mean-field model closer ro reality. Finally, it should be pointed out that for the NH_3 experiments cited, extremely low pump powers were utilized and only resulted in small modifications to the $\tilde{\gamma}$ in these experiments.

Without new theoretical and experimental work towards these goals, there is little distinction between the recent experimental efforts and the early observations of self-pulsing in homogeneously broadened optically pumped molecular lasers [23,24]. Finally it should be pointed out that the lack of agreement between parameter predictions and actual experiments in dynamical systems has plagued many fields [25]. Interestingly enough, what were believed to be only quantitative disagreements have led to new classes of qualitative behavior and a much greater understanding of the problems under study.

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