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Publication Date

1975-06-01

0 0 0 0 4 3 0 5 3 2 3

Presented at the Proceedings of the
27th International Meeting of the
Societe de Chimie Physique, Paris, France,
June 17 - 20, 1975

LBL-3916
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C. B. Harris

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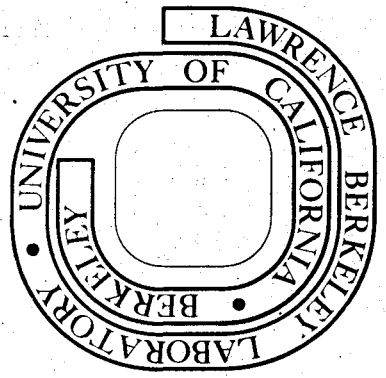
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THE FEASIBILITY AND ADVANTAGES OF OFF-RESONANCE LASERS IN CHEMICALLY
REACTING SYSTEMS

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SUMMARY

The problem of understanding the semiclassical description of the time evolution of an ensemble of two state systems under the influence of a coherent radiation field is of considerable importance. Previous attempts to deal with these problems have dealt with either broad pulses or ultrashort pulses which allow the use of the rate equations or finite phase memory to be incorporated into the description. In neither case, however, has the effect of incoherent feeding and off-resonance effects in a coherently driven two-level system been analyzed. A closed form solution that includes the effects of relaxation and spontaneous emission between the two levels has been obtained for the general case when the ensemble is being incoherently fed from a population reservoir, as would be the case for example in a chemical laser. In addition to providing a basis for understanding the modifications which occur for such a system, the mathematical formulation predicts that an important effect may be observed. This effect, which is termed "kinetic coherence," is the production of a long-term coherent component that results directly from the kinetic feeding. The magnitude of the component is related to the rate of creating excited states, relaxation pathways and the off-resonance frequency. It will be shown how in principle it is possible to utilize these off-resonance effects in any inhomogeneously broadened system to significantly overcome the losses from T_2 relaxation processes and to provide an experimental system capable of controlling the relative ratio of spontaneous and stimulated emission. Finally, the relationships between chemical kinetics, the off-resonance feature and sustained self-regulation in a system exhibiting gain will be discussed.

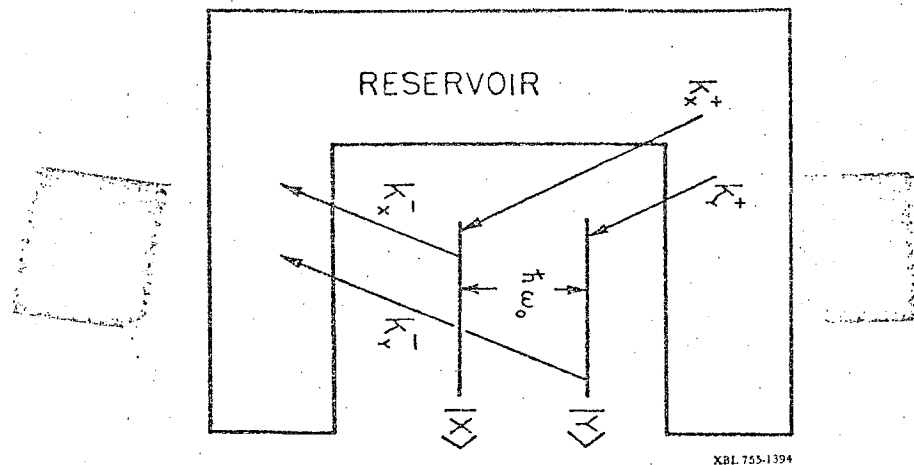
INTRODUCTION

The problem of understanding the time evolution of an ensemble of two-level systems under the influence of a coherent radiation field is basic to a variety of fields in optics and in magnetic resonance. In almost all cases, a semiclassical description of the interaction between the particles and radiation has been used [1-3] and the set of coupled differential equations has been solved by the constraints of the strong-field short pulse approximation [4] or, as is done in magnetic resonance, by assuming the total population in the ensemble is constant in time [5]. Little attention has been given to examples where the total population of the two-level system is time-dependent. In some respects the work of Icsevgi and Lamb [6] on pulse propagation in laser amplifiers comes closest to addressing itself to this problem; however they did not explicitly formulate the qualitative and quantitative effects that "kinetic" processes in the ensemble of excited states play in the creation of the coherent superposition of states. In this paper, the role that incoherent feeding and decay play in the production of coherent states will be outlined. Particular attention will be given to the implications of off-resonance fields as a means of generating steady state coherence in the ensemble that results directly from a combination of the incoherent feeding process and the driving field. This effect has been termed "kinetic coherence" [7], and under certain conditions can be several orders of magnitude larger than the usual on-resonance component. Because of the time and space limitations in these proceedings, the development will be outlined only. A complete description of the problem, however, will be published elsewhere [7].

FORMULATION OF THE PROBLEM

(1) Generality of the Magnetic and Optical Case

In Figure 1 the salient rate processes to be considered are diagrammatically illustrated. k_x^+ and k_y^+ represent the incoherent feeding rates into the two-level system $|x\rangle$ and $|y\rangle$, respectively, while k_x^- and k_y^- represent the decay rates in the absence of a driving field. The Hamiltonian responsible for the coherent radiation field, $V(t)$, is applied at a frequency ω which may or may not be at the center frequency, ω_0 . The model for feeding and decay contains the following assumptions [7]:



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Fig. 1: Diagrammatic representation of a two-level system coupled by a coherent radiation field in the presence of feeding rates into the individual levels k^+ , and decay rates from the two levels k^- .

(a) Feeding only occurs to eigenstates of the ensemble of two-level systems and not to coherent superposition states; however, decay occurs from both the eigenstates and from superposition states depending upon the explicit effects of the driving field. (b) The feeding rates into the eigenstates are constant and independent of the state of the ensemble. (c) The decay rate of the ensemble depends on the state of the ensemble and consequently the total population need not be time or field independent. This model is simple, and approximates many physically realizable situations.

Two general cases can be distinguished at this point. The first is representative of magnetic oscillating dipole fields and is formulated by allowing the coherent driving field to be of the form,

$$V(t) = \gamma \mathcal{C}_1 \hat{S}_1 \cos(\omega t + \phi) \quad [1]$$

$\gamma \mathcal{C}_1$ is the strength of the oscillating field applied at a phase ϕ . Solutions of the coupled differential equations including feeding and decay are applicable in this case to a variety of problems in magnetic resonance, including triplet state spin dynamics [8,9], optically pumped electron and nuclear spin polarization [10], chemically induced nuclear and electron spin polarization [11,12], spin diffusion, and others where the wavelength of the driving field is greater than the sample size, i.e., $\lambda^3 \gg \text{volume}$. Without loss of generality, one can consider cases where the wavelength of the radiation field is small relative to the size of the sample (the optical region) and the coherent Hamiltonian is an

oscillating electric dipole field:

$$V(t) = -\mu \cdot E_1 \cos(\omega t + \phi - \vec{k} \cdot \vec{r}) \quad [2]$$

\vec{k} is the wavevector of the radiation field and μ is the transition dipole moment associated with $|y\rangle \rightarrow |x\rangle$. If one considers "thin samples," the coupled Maxwell and Schrödinger equations may be avoided, thus allowing an exact solution to be obtained for both steady state and transient behavior of the ensemble in the presence of feeding and decay for all strengths of the driving field and in the presence of phenomenological relaxation terms [13]. This is true for either form of the time-dependent Hamiltonian in Eqs. 1 and 2. Indeed, one can show that the equation of motion for the time-dependent density matrix associated with Eqs. 1 and 2 is of the same form when the explicit time dependence is removed from Eq. 1 and the explicit time and spatial dependence is removed from Eq. 2. The appropriate transformations are

$$U^{-1} = \exp(i\mathcal{H}_0 \omega t / \hbar \omega_0) \quad [3]$$

and

$$U^{-1} = U_{k,j}^{-1} = \exp[i\mathcal{H}_0 (\omega t - \vec{k} \cdot \vec{r}_j) / \hbar \omega_0] \quad [4]$$

respectively. The resulting time-independent density matrix in the resulting rotating frame is given by

$$\rho^*(t) = U \rho(t) U^{-1} \quad [5]$$

Both Eqs. 3 and 4 satisfy the equation of motion, $i\mathcal{H}\rho^* = [\mathcal{H}^*, \rho]$, where

$$\mathcal{H}^* = U[\mathcal{H}_0 + V(t)]U^{-1} \quad [6]$$

and \mathcal{H}_0 is the zeroth order Hamiltonian having eigenvalues of $\pm \hbar \omega_0 / 2$.

In the optical regime, solutions to the coupled differential equations serve as a potential model for understanding a variety of kinetic problems including chemical laser dynamics [14,15], stark shift optical coherence [16], pulse propagation [6,17] in an amplifying media, to mention only a few.

(2) Outline of the Solution

There are at least two distinct ways of solving the coupled differential equations. The first [7] is to formulate the density matrix in such a way that it operationally includes both processes. Specifically, the density matrix describing a two-level system restrained by assumptions (a)-(c) above is

$$i\hbar \frac{d\rho}{dt} = [\mathcal{H}, \rho] - [K, \rho]_+ + F \quad [7]$$

where the feeding terms are given by

$$F = i\hbar \begin{bmatrix} k_y^+ & 0 \\ 0 & k_x^+ \end{bmatrix} \quad [8]$$

and the decay terms are incorporated by constructing an imaginary operator:

$$K = \frac{i\hbar}{2} \begin{bmatrix} k_y^- & 0 \\ 0 & k_x^- \end{bmatrix} \quad [9]$$

The final solution to Eq. 7 can be shown [7] to be

$$\rho(t) = Q^\dagger [\rho(0) - \rho_s] Q + \rho_s \quad [10]$$

where ρ_s is the density matrix describing the steady state to which the ensemble evolves under a given set of experimental conditions. Q is given by

$$Q = \exp\{i[(\mathcal{H} + K)/\hbar]t\} \quad [11]$$

The second method [18] results in an expression identical to the above, but is more cumbersome to develop mathematically. It retains, however, considerably more physical insight into the problem. In brief, the problem is solved by incorporating the kinetic processes directly into the time-dependent Schrödinger Equation by subpartitioning the ensembles of two-level systems properly in time as not to disregard information concerning the relative phases of the states which are continually being

created. One first solves the coupled differential equations for the initial $t = 0$ population, including the effect of decay and then separately solves the equation of motion for those additional states that are created in a small increment of time, δt . At any time t the solution is simply the initial solution $\rho_0(t)$, plus $\rho_{\delta t}(t) + \rho_{2\delta t}(t) + \dots + \rho_{n\delta t}(t)$, where $t = n\delta t$ and $\rho_0, \rho_{\delta t}, \rho_{2\delta t}$, etc., are the $t = 0, t = 0$ to $\delta t, t = \delta t$ to $2\delta t, \dots, t = (n-1)\delta t$ to $n\delta t$ subensembles, respectively. In the limit that $\delta t \rightarrow 0$ one obtains a set of solvable coupled integral equations. The analytic solutions [7] are formidable and will not be presented here; however, the qualitative features of the results are simple and revealing, particularly in the rotating frame.

EFFECTS OF FEEDING INTO A COUPLED TWO-LEVEL SYSTEM

(1) On-Resonance Feeding

Consider first on-resonance driving fields in the case where the ensemble is initially inverted, i.e., all the population is in the upper level $|y\rangle$ at $t = 0$, as illustrated in Figure 2. When the driving field is externally turned on as in magnetic resonance or internally builds up from the reactance field according to Maxwell's Equation as in an optical oscillator or amplifier, the ensemble is driven around the field. In the case of an oscillating magnetic dipole, it executes a transient nutation and dephases because of relaxation. In the absence of feeding at some later

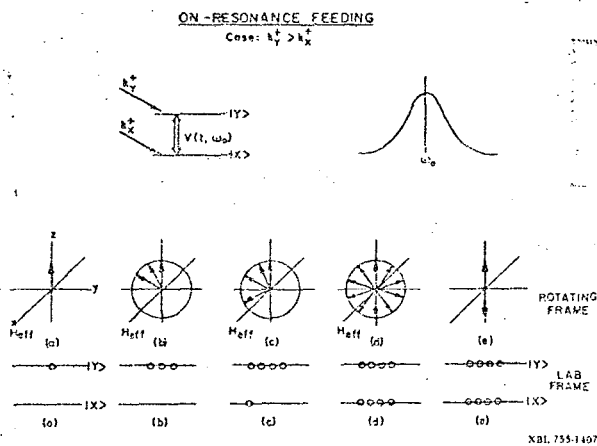


Fig. 2: The relationship between the rotating and laboratory frames on-resonance in the presence of feeding processes.

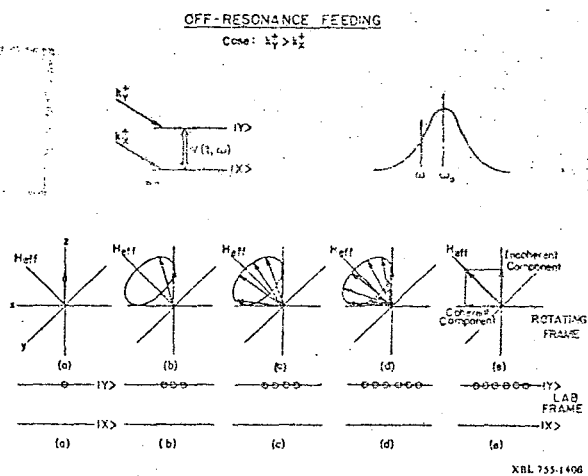


Fig. 3: The relationship between the rotating and laboratory frames off-resonance in the presence of feeding processes.

time, $\rho_0(t)$, the density matrix describing the initial population contains a random and equal distribution of vectors in the yz plane of the rotating frame, x being associated with $\phi = 0$ of the driving field. In the optical case for a monochromatic wave [6], the reactance field will drive the ensemble to saturation, causing the gain to reach a limiting value. Naturally transient oscillations in the reactance field are possible as is the case with optically induced self-transparency [19]. In the case of an amplifier the limiting steady state intensity approaches a value that is determined by the amount by which threshold is exceeded and by the Lorentz profile for homogeneous relaxation. As we will see, such is not the case for the off-resonance driving field.

When one considers the feeding processes as illustrated in Fig. 2, the ensemble also evolves to a saturated value for on-resonance fields. This can be seen by considering the case where $k_y^+ \gg k_x^+$. In such instances, excited states (subensembles) are incoherently created in time, each along the $+z$ axis in the rotating frame because there is no phase relation between the excited states that are created and those that already exist in the ensemble. Each subensemble is in turn driven by the field in the yz plane. In both the optical and magnetic cases, a steady state saturated value is reached, and is represented as a disk of vectors in the yz plane, each vector representing one of the subensembles. Thus, for on-resonance feeding, the driving field equally and incoherently distributes the population into the two levels, $|x\rangle$ and $|y\rangle$. The only exception to this would be if the decay rates were much shorter than the transient nutation frequency in the yz plane. In such a case for a magnetic dipole, a steady state polarization would exist that generates a coherent component 90° out of phase with the driving field. In the optical case, however, the reactance field would presumably accelerate the ensembles to such an extent that it might be difficult to have the nutation rate slower than the decay rate at steady state.

(2) Off-Resonance Feeding

When the driving fields are applied, or develop off-resonance at a frequency ω by an amount $\Delta\omega$, given by

$$\Delta\omega = \omega_0 - \omega, \quad [12]$$

several new phenomena result from the kinetic feeding and decay processes. Consider the case where feeding into one of the two levels is preferred

($k_x^+ \gg k_y^+$). In such instances the $t = 0$ ensemble will precess about an effective field which is at some angle in the xz plane determined by $\Delta\omega$. Because the states are radiatively or nonradiatively decaying at k_x^- and k_y^- , the initial polarization will be lost after a time comparable to $(k_x^- + k_y^-)^{-1}$. The states that are being continually created by k_y^+ , however, enter randomly along $+z$ at a rate proportional to k_y^+ , and like the $t = 0$ subensemble, decay as k_x^- , k_y^- or some average value. If the feeding rate is comparable to the decay rates and the driving field is sufficiently large, then a cone of vectors will develop around the effective field. This cone will evolve to some steady state value depending upon the magnitude of various relaxation and kinetic processes. This is illustrated in Figure 3.

The important qualitative points to note are: (i) a steady state nonsaturated value for the polarization is achieved by kinetic feeding; (ii) this polarization appears "locked" on the field; (iii) relaxation is overcome when H_{eff} is large enough because the off-resonance condition is satisfied for all members of a Lorentz absorption regardless of whether the transition is homogeneously or inhomogeneously broadened; (iv) there is a coherent component of the polarization which is simply the projection of the steady state value of the lock polarization onto the xy plane. This has been called kinetic coherence because its magnitude results directly from the kinetic parameters; and finally, (v) the coherent component can be stabilized and maximized when the off-resonance frequency is chosen properly.

The off-resonance value, $\Delta\omega_{\text{max}}$, that corresponds to a maximum coherent component $r_1^s(\text{max})$ is given by [7]:

$$\Delta\omega_{\text{max}} = 1/T_2^* + (1/\sqrt{T T_e})(1 + \omega_1^2 T T_e)^{1/2} \quad [13]$$

and yields a value for the Feynman, Vernon and Hellwarth [20] r_1 component of

$$\overline{r_1^s}(\text{max}) = [r_3^0 \omega_1 / 2] [1/T_2^* + (1/\sqrt{T T_e})(1 + \omega_1^2 T T_e)^{1/2}]^{-1} \quad [14]$$

where T_2^* is the inhomogeneous relaxation time associated with a Lorentz lineshape function, $g(\omega_0)$;

$$g(\omega_0) = [T_2^*/\pi] [1 + (\omega - \omega_0)^2 T_2^{*2}]^{-1} \quad [15]$$

where

$$\int_{-\infty}^{+\infty} g(\omega_0) d\omega_0 = 1 \quad [16]$$

and T_e contains relaxation along the field [21] T_{2e} as

$$1/T_e = [(k_x^- + k_y^-)/2] + T_{2e}^{-1} \quad [17a]$$

T and τ contain the kinetic parameters and relaxation terms. T is given by

$$1/T = [(k_x^- + k_y^-)/2] + 1/T_2 \quad [17b]$$

where T_2 is the normal transverse relaxation time. τ is given in terms of the kinetic parameters and T_1 processes as:

$$\tau = [(k_x^- + k_y^-)/2] [k_x^- k_y^- + k_y^- T_x^{-1} + k_x^- T_y^{-1}]^{-1} \quad [18]$$

T_x and T_y allow for both spontaneous emission from $|y\rangle$ to $|x\rangle$, T_{is} , and normal T_1 type terms, T_{li} :

$$T_i^{-1} = T_{is}^{-1} + T_{li}^{-1} \quad (i = x \text{ or } y) \quad [19]$$

The relationship between the feeding process and the coherent components in Eq. 14 is given by r_3^0 , which is [7]:

$$r_3^0 = [\tau / (k_y^- + k_x^-)] [(k_y^+ k_x^- - k_x^+ k_y^-) + (k_y^+ + k_x^+) / T_x - (k_y^+ + k_x^+) / T_y] \quad [20]$$

Finally, ω_1 is the strength of the driving field and is given by the strength of the transition dipole. For the magnetic case it is

$$\omega_1 = \gamma \mathcal{C}_1 F \quad [21]$$

while for the optical case it is

$$\omega_1 = \mu \cdot E_1 \quad [22]$$

When the driving field strength is large, i.e.,

$$\omega_1 T_2^* > 1 \quad [23]$$

$$\omega_1 T > 1 \quad [24]$$

and

$$\omega_1^2 T \tau > 1, \quad [25]$$

the maximum kinetic coherent component is given by a simple expression:

$$r_1^S(\text{max}) = (r_3^0/2)(\sqrt{T_e/\tau}) \quad [26]$$

while for on-resonance, the coherence is given by [7]:

$$r_2^S = [-r_3^0 \omega_1] [\omega_1^2 \tau + \{1/T + \sqrt{T_e/T} T_2^{*-1} (1 + \omega_1^2 T \tau)^{1/2}\}]^{-1} \quad [27]$$

The important point to note is that the maximum value on-resonance for an inhomogeneous transition is significantly less than possible values off-resonance. This is illustrated in Table 1, where the ratio of on-resonance to off-resonance coherence is given versus a measure of the homogeneous to inhomogeneous linewidth ratio, T/T_2^* . For example, (cf. Table 1) if a transition is inhomogeneously broadened by hundredfold the homogeneous width, then the maximum coherence on-resonance is less than one percent of the value that could be obtained off-resonance. Therein lies the central importance of off-resonance driving fields.

Table 1: Ratio of off-/on-resonance coherence versus T/T_2^* in units of r_3^0 (see Eq. 20).

T/T_2^*	0	1	10	100
Off-/on-r.c.	0.5/0.5	0.5/0.3	0.5/0.075	0.5/0.009
$\Delta\omega(\text{max})$ in units of T_e^{-1} (Eq. 17a).	~2.5	~2.5	~2.5	~2.5

APPLICATION TO CHEMICAL LASERS

In some respects chemical lasers [14] are ideally suited to test the above theory and predictions. Many of the inherent limitations that are associated with chemical lasers and chemically reacting systems might be

circumvented by the development of "off-resonance" type lasers.

An oversimplified picture of chemical lasers is that a nonequilibrium distribution of final product states is created by selective channeling of energy from a chemical reaction in such a way that a partial population inversion is created between the molecular rotation-vibration states of the products. In this sense the kinetic parameters k_y^+ and k_x^+ in the above description can analytically incorporate the chemical kinetics associated with the formation of products into particular vibrational-rotation states. In the absence of vibrational relaxation, the population of the various states, N_{VJ} , is proportional to microscopic rate constant k_V for product formation [22] and hence determines the extent of population inversion in the various P-branch ($\Delta J = + 1$) and R-branch ($\Delta J = - 1$) transitions.

One can go quite far in the analogy between chemical lasers and the above model. It is well-known that relaxation processes usually limit the storage of energy in excited states, insofar as stimulated emission competes with collisional deactivation processes, vibration deactivation being the principal shortcoming of most hydrogen halide lasers [23,24]. The details of vibrational, rotational [25] and other relaxation processes are contained in the relaxation terms, k_y^- , k_x^- , T_{1x} and T_{1y} . Consider, for example, a single P-branch transition in the HF laser, $P_2(3)$. In such cases, the levels $|y\rangle$ and $|x\rangle$ are associated with the product states ($V = 2, J = 2$) and ($V = 1, J = 3$) in HF respectively. k_y^+ and k_x^+ would be related to the bimolecular rate constants k_2 and k_1 for the formation of HF ($V = 2$) and HF ($V = 1$) as:

$$k_y^+ = k_2 \quad [28]$$

$$k_x^+ = k_1 \quad [29]$$

Vibration relaxation to all levels except $V = 1, J = 3$ from $V = 2, J = 2$ is given by

$$k_y^- = \sum_{V \neq 1; J \neq 3} \sum P(2,2|VJ) \quad [30]$$

where $P(i,j|k,\ell)$ are proportional to the probabilities for the individual vibration-rotation decay channels. Likewise,

$$k_x^- = \sum_{V \neq 2} \sum_{J \neq 3} P(1,3|VJ) \quad [31]$$

Finally, $P(2,2|1,3)$ and $P(1,3|2,2)$ are proportional to T_{1x}^{-1} and T_{1y}^{-1} , respectively, in Eqs. 18 and 19. In this manner, most of the salient rate processes are incorporated into a single unified description.

At this point two limiting cases should be distinguished to illustrate the potential advantages and usefulness of off-resonance effects. These are the low and high pressure regimes which are characterized by an inhomogeneous or homogeneous transition, respectively.

At low pressure (< 1 torr) collisions are infrequent enough on the time scale of the stimulated emission output (μsec) that there is no significant equilibration of the translation kinetic energy. To a first approximation this limits laser action to regions of the Doppler profile that overlaps the cavity modes. This severely limits the number of molecules that can contribute to the stimulated emission gain and hence the output of the laser on-resonance is reduced by approximately (nT_2^*/T_2) , where n is the number of cavity modes that reach threshold and $(T_2^*/T_2)^{-1}$ is the ratio of the inhomogeneous to homogeneous linewidth. For low enough pressures this reduction can be ten to hundredfold or more. Low pressures, however, have important advantages because the vibrational (and/or rotational) deactivation is significantly reduced. This in principle can result in much larger inversions, particularly for the higher vibrational states. Unfortunately this has not been realized operationally in the normal use of most chemical lasers because of the necessity of having a sufficient number of collisions to homogeneously (pressure) broaden the line in order to obtain a full inversion under the entire line profile. An off-resonance laser overcomes this limitation insofar as the full inhomogeneous distribution contributes to stimulated emission independent of collisional broadening. This would mean that in principle the entire inhomogeneous profile in the state of highest inversion could contribute to stimulate emission. If the effects of spontaneous emission on-resonance were minimized in the time necessary to reach threshold off-resonance, the maximum gain from the system could be realized. In addition, because the gain/molecule is low off-resonance, off-resonance lasers might be ideal low noise amplifiers. On-resonance spontaneous emission in the mode structure of the cavity, however, would have to be absorbed or limited.

Finally, I wish to consider qualitatively at least the advantages "off-resonance" chemical lasers potentially have at higher pressures (50-100 torr or greater). Chemical lasers operating at higher pressures suffer from the competition between stimulated emission, and vibrational and rotational deactivation. Although this is currently thought to limit their applicability as high power oscillators or amplifiers, TEA lasers have demonstrated that with a rapid enough build-up in the population inversions, reasonable powers can be achieved in some systems operating at pressures as high as atmospheric [26]. In such cases the line can be considered homogeneously broadened on a nanosecond time scale because of the very rapid equilibration between the Doppler components of transition. Translational equilibration occurs on a time scale given by the collision frequency at room temperature and atmospheric pressure (~ 4 collisions/nsec) [27].

Although the maximum off-resonance and on-resonance coherence are the same for a given set of rate processes (cf. Table 1), the two classes differ with respect to the strength of the driving field necessary to achieve the optimal coherence. For the on-resonance case the coherent component is always higher than that of off-resonance until saturation is reached. For conventional low powered lasers "off-resonance" operation in the homogeneous regime offers little advantage. For higher powers, however, on-resonance systems begin to "power broaden" so that the effective field strength can be significantly reduced. This can be seen by the factor of $\omega_1^2 \tau$ in the denominator of Eq. 27. In the off-resonance mode, however, no power broadening is predicted to occur, so that the maximum coherence can always be obtained in the higher power limit without the complication of saturation and subsequent field dependent broadening.

ACKNOWLEDGEMENTS

This research was supported in part by the National Science Foundation and in part by the U.S. Energy Research and Development Administration.

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