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Cooperative Relaxation in Coherently Pumped Three-Level Systems

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The behavior of a large number of coherently pumped three-level systems coupled to two resonant cavity modes at the two respective intermediate frequencies is shown to be qualitatively different from conventional laser-type behavior. The existence of three steady states and the dependence of their stability on the operating conditions allows the production of steady, modulated, or pulsed excitation of both modes.

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Coherent pumping of lasers (by other lasers) has become common practice, and pump lasers are available in a wide range of frequencies. The operation of a three-level laser depends both on induced emission at one intermediate frequency and on incoherent relaxation at the other. These two processes may be regarded as cooperative and noncooperative relaxation, respectively. It is the present purpose to analyze a novel and interesting type of possible behavior—qualitatively different from conventional laser-type behavior—in which the three-level atomic systems (hereafter referred to as "molecules") undergo cooperative relaxation at *both* intermediate frequencies. This may be accomplished by letting molecules for which all three transitions couple to the electromagnetic field interact with two (lossy) cavity modes tuned to the two respective intermediate frequencies. Labeling the three molecular energy levels in ascending order by $\hbar\omega_i$, $i=1, 2, 3$, we consider a model in which N identical molecules are coupled to two modes with respective frequencies ω_{12}, ω_{23}, and pumped at frequency ω_{13}, where $\omega_{ij}=|\omega_i-\omega_j|$. While such coupling, in accordance with well-known selection rules, may be too weak in most atomic systems to produce the present effects, there exist atoms and molecules for which forbidden lines or overtones are sufficiently strong (such as the OCS molecule[1]) or for which two-photon pumping may be possible. The present discussion is not necessarily restricted to optical or infrared frequencies but is also applicable to microwave frequencies.[2]

In order to exhibit most simply the new feature, we introduce the idealizations that the coupling strength between molecule and mode depends only on the mode, and that transitions other than those due to induced emission are negligible. The former is used widely in analyses of cooperative phenomena,[3] and the latter will be discussed further. We use a formalism which is especially suitable for the analysis of cooperative phenomena and can be read directly both quantum mechanically and classically.[4,5] The atomic equations of motion, in the rotating-wave approximation, are given in this formalism by

$$\dot{A}_1=\omega A_3-i\gamma_{12}B_{12}^\dagger A_2,$$
$$\dot{A}_2=-i\gamma_{23}B_{23}^\dagger A_3-i\gamma_{12}A_1B_{12},$$
$$\dot{A}_3=-\omega A_1-i\gamma_{23}A_2B_{23}.$$

Briefly, the A's and B's are the variables that describe the collection of molecules and the cavity fields, respectively, and are related to a_i ($a_i^\dagger$), the annihilation (creation) operator of atoms in the ith level, and to b_{ij} ($b_{ij}^\dagger$), annihilation (creation) operators of photons in the ij mode, respectively, by $a_j=A_j\exp(-i\omega_j t)$, $b_{jk}=B_{jk}\times\exp(-i\omega_{jk}t)$; ω refers to the pump field, 2ω being the Rabi frequency of the (1,3) pair of levels, and the γ's are real coupling constants.[6] The equations are consistent with the normalization $\sum A_i^\dagger A_i=N$. Since our interest lies in macroscopic phenomena, with N large, the classical description is a good approximation under most conditions,[4] and will be used henceforth. We consider only the case of sufficiently damped cavity modes so that the fields follow the respective resonant polarizations adiabatically, as expressed by[7] $B_{jk} \approx -i(\gamma_{jk}/\xi_{jk})A_j^{\dagger}A_k$, where $2\xi_{jk}$ is the energy relaxation constant of the jk mode.

Eliminating the field from the molecular equations of motion, and setting $x_i = A_i/N^{1/2}$, $\tau = \omega t$, $c_{ij} = N\gamma_{ij}/\xi_{ij}\omega$, one obtains

$$x_1' = x_3 + c_{12}x_1|x_2|^2,$$
$$x_2' = c_{23}x_2|x_3|^2 - c_{12}|x_1|^2x_2,$$
$$x_3' = -x_1 - c_{23}|x_2|^2x_3,$$

the prime indicating differentation with respect to τ. Clearly, the phase of x_2 is constant, and is given by its initial value. Elimination of $|x_2|^2$ yields

$$x_1' = x_3 + c_{12}x_1(1 - |x_1|^2 - |x_3|^2),$$
$$x_3' = -x_1 - c_{23}x_3(1 - |x_1|^2 - |x_3|^2).$$

The most interesting solutions, from a physical viewpoint, are included in those for real x_1 and x_3, for the following reasons. If $x_1(0)$ and $x_3(0)$ are real, $x_1(\tau)$ and $x_3(\tau)$ will be real. Either $x_1(0)$ or $x_3(0)$ can be chosen real, since only phase *differences* are physically significant. If $x_1(\tau)$ or $x_3(\tau)$ vanishes, the nonvanishing variable may be redefined to be real, and both variables will continue to be real henceforth. For steady states, choosing one variable to be real makes the other real, and the same applies if only one derivative vanishes at any time. We will confine our attention, therefore, to real values of x_1 and x_3. For notational simplicity, we set $x_1 = x$, $x_3 = y$, $c_{12} = c_1$, $c_{23} = c_2$ and obtain, as equations of motion

$$x' = y + c_1x(1 - x^2 - y^2),$$
$$y' = -x - c_2y(1 - x^2 - y^2),$$

which, in polar coordinates (with $x = r\cos\theta$, $y = r\sin\theta$) become

$$r' = r(1 - r^2)(c_1\cos^2\theta - c_2\sin^2\theta),$$
$$\theta' = -1 - \tfrac{1}{2}(c_1 + c_2)(1 - r^2)\sin 2\theta.$$

There exist two steady-state solutions, which we label Z and S, respectively, Z being the solution $r = 0$, and S being the solution $r^2 = 1 - (c_1c_2)^{-1/2}$, $\tan\theta = -(c_1/c_2)^{+1/2}$, with S existing only for $c_1c_2 > 1$. There exists another solution with some steady-state properties, which we label p: $r = 1$, $\theta = -\tau$. For convenience, all three will be referred to as steady states. (In the terminology of nonlinear equations,[8] Z and S are singular points, and P is a limiting cycle.) The stability condition of the steady states (in the sense of "asymptotic orbital stability"[8]) are as follows: for Z, $c_1/c_2 < 1$ and $c_1c_2 < 1$; for S, $c_1/c_2 < 1$ (and $c_1c_2 > 1$, of course); for P, $c_1/c_2 > 1$. The stability regions in the c_1-c_2 plane are illustrated in Fig. 1.[9] The only steady state in which the cavity modes are excited is S. In Z, the total population is in the middle level, while in P, all molecules undergo a Rabi oscillation between first and third levels.

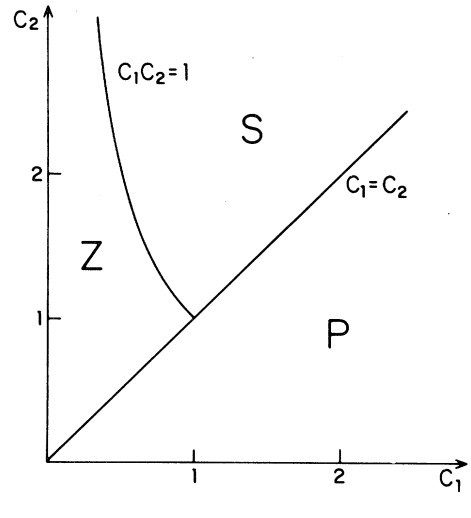

FIG. 1. Regions of stability in the c_1, c_2 plane for the three steady-state solutions.

The most interesting behavior is exhibited by the time-dependent solutions. For $c_1 = c_2 = c$, analytic expressions for the trajectories corresponding to these solutions are given by

$$1 - r^2 = K\exp(cr^2\sin 2\theta)$$

where K is a constant of integration determined by the initial conditions. This equation describes a family of closed curves as shown schematically in Fig. 2, each curve corresponding to a different K. For $c_1 \neq c_2$, one can see from the polar-coordinate form of the equations of motion that the trajectory representing the solution will spiral in or spiral out (if the initial state is not a steady state), depending on whether $c_2 > c_1$ or $c_1 > c_2$, approaching the stable steady state while following roughly the outline of the periodic trajectories of Fig. 2. Two computer graphs of such solutions are shown in Fig. 3. The field energy in the (1,2) cavity is given by $\hbar\omega_{12}(\gamma_{12}/\xi_{12})^2N^2x^2(1 - r^2)$ and that in the (2,3) cavity by $\hbar\omega_{23}(\gamma_{23}/\xi_{23})^2N^2y^2(1 - r^2)$, so that the fields are modulated (with opposite phase, when the trajectory spirals around the origin).

A sudden change in ω, that is, in the pump strength, for $c_1 < c_2$, which produces a crossing

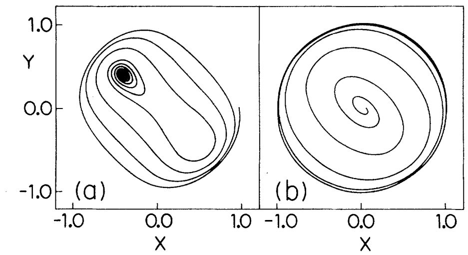

FIG. 2. Trajectories in the x-y plane for $c_1 = c_2 = c$. (a) $c = 1.5$. (b) $c = 0.5$. Each closed curve represents the solution for a given $x(0)$, $y(0)$ (or a given value of K). The outer circle is solution P, the center is solution Z, and the two off-center points in (a) are solution S. Dotted curve is separatrix.

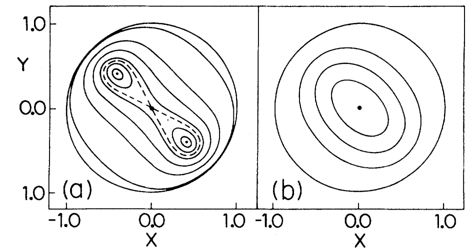

FIG. 3. Trajectories in the x-y plane for $c_1 \neq c_2$. (a) $c_1 = 1.4$, $c_2 = 1.6$, $x(0) = 0.95$, $y(0) = 0$. (b) $c_1 = 0.7$, $c_2 = 0.4$, $x(0) = 0.05$, $y(0) = 0$.

of the curve $c_1c_2 = 1$ (see Fig. 1), will produce a transition from S to Z (a modulated decay of the fields) for an increase in pump strength, and a transition from Z to S (a modulated rise of the field) for a decrease in pump strength. The $S \rightarrow Z$ transition is caused by the disappearance of S, while the $Z \rightarrow S$ transition is caused by an (assumed) arbitrarily small perturbation of the (unstable) Z state. A particularly interesting effect is the occurrence of a modulated pulse in both modes as a result of a $Z \leftrightarrow P$ transition, when stability is shifted from Z to P or vice versa by crossing the line $c_1 = c_2$ (a line of "bifurcation points"[8]). Such a transition can be produced by a change in ξ_{12} or ξ_{23}, the cavity loss, conceivably by electronic methods. The $Z \rightarrow P$ transition is illustrated in Fig. 3b (for a perturbed Z); the $P \rightarrow Z$ transition is a spiraling in from $r = 1 - \epsilon$ to $r = 0$. Note that a period of revolution along the spiral is approximately a Rabi period $\pi\omega^{-1}$, and the pitch of the spiral (near a given r) varies as $c_1 - c_2$. A $P \rightarrow S$ transition is illustrated in Fig. 3a. Several other types of time dependent solutions will be discussed elsewhere.[10]

Atomic relaxation and spontaneous emission have been ignored. These will produce the perturbations of the unstable steady states that result in some of the transient pulses described above, and perturbations of the stable steady states that are of little qualitative significance for sufficiently strong pumping.[11] One may reasonably expect that after the begining of a transient mode excitation the induced emission will become dominant and other relaxation effects relatively negligible for sufficiently short pulses.

An insight into the fundamental difference between the present phenomena and conventional laser-type effects is furnished by the following characteristics of the present system: 1) If a coherently oscillating dipole moment exists with respect to one pair of intermediate levels, it must also exist with respect to the other pair, so that both modes must oscillate together. 2) Since the main relaxation effect is determined by the mode damping, it can be controlled simply, allowing the shift of stability from one steady state to another and the production of the associated transition pulses. 3) Quasisteady-state oscillation can occur (for $c_1 = c_2$) in which the energy in the two modes undergoes modulation with opposite phase for constant pump field.[12]

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[1]J. Braslawsky and Y. Ben-Aryeh, J. Mol. Spectrosc. 30, 116 (1969), and references therein. For the bending mode of the OCS molecule ($\nu_2 \approx 527$ cm^{-1}), the dipole moments associated with excitation of the fundamental and the overtone are both ~ 0.1 D. Another example of comparable dipole moments for the fundamental and overtone is furnished by the bending mode of the HCN molecule ($\nu_2 \approx 712$ cm^{-1}).

[2]The fact that three frequencies—with one being the sum of the other two—are involved should not lead to an identification of the present system with a parametric oscillator. The latter consists, essentially, of three coupled oscillators, one of which is the pump [see, for instance, D. F. Walls and R. Barakat, Phys. Rev. A 1, 446 (1970), and I. R. Senitzky, Phys. Rev. 183, 1069 (1969)], while the former consists of three resonant *pairs* of oscillators (including the pump). The number of degrees of freedom of the present system is larger than that of the parametric oscillator.

[3]For a large number of references, see *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New

York, 1977).
[4]I. R. Senitzky, Phys. Rev. A 10, 1868 (1974), and 15, 284 (1977).
[5]I. R. Senitzky, in *Progress in Optics*, edited by Emil Wolf (North Holland, Amsterdam, 1978), Vol. 16.
[6]The cooperative molecular amplitudes a_i can be related to a more familiar formalism in the case of a collection of *two-level* systems. In this case the collective (dimensionless) angular momentum operators l_1, l_2, l_3 are given by $l_1 = \frac{1}{2}(a_1 a_2^\dagger + a_1^\dagger a_2)$, $l_2 = -\frac{1}{2}i(a_1 a_2^\dagger - a_1^\dagger a_2)$, $l_3 = \frac{1}{2}(a_2^\dagger a_2 - a_1^\dagger a_1)$. Cooperation corresponds to the invariance of l^2. The present model is subject, of course, to the same limitations as the model for a collection of two-level systems in which l^2 is invariant. The latter model and its limitations are discussed by I. R. Senitzky, Phys. Rev. A 6, 1171 (1972), and by P. D. Drummond and H. J. Carmichael, Opt. Commun. 27, 160 (1978).
[7]I. R. Senitzky, Phys. Rev. A 15, 292 (1977), Eqs. (2.19) and (2.22), and Phys. Rev. 155, 1387 (1967), Sec. I.
[8]H. Haken, *Synergetics* (Springer, Berlin, 1977), Chap. 5.
[9]The similarity of Fig. 1 to a phase diagram is obvious. If $|x_2|$ is considered (formally) to be the "order parameter", then the transition $S \leftrightarrow Z$ corresponds to a second-order phase transition, while $S \leftrightarrow P$ and $Z \leftrightarrow P$ correspond to first-order phase transitions. Nonequilibrium phase transitions involving cooperative atomic behavior of two-level systems are discussed by D. F. Walls, P. D. Drummond, S. S. Hassan, and H. J. Carmichael, Prog. Theor. Phys. (Japan). Suppl. 64, 307 (1978), who also give additional references.
[10]Oscillatory approaches to equilibrium are found also in driven cooperative two-level systems. See L. Narducci *et al.*, Phys. Rev. A 18, 1571 (1978), and papers in Ref. 6. That of Drummond and Carmichael contains additional recent references.
[11]The apparent—seemingly puzzling—stability of P and S in the limit $\omega \to 0$ is due to the fact that relaxation in the (1, 3) transition has been ignored. The present model is applicable only when the effect of this relaxation is small compared to that of the pump. Otherwise, sufficiently small ω leads to a fourth stable state, to be described in detail elsewhere, which is irrelevant to the effects presently considered. With a more complex model (beyond the scope of the present discussion), it can be shown that this state is a weakly excited steady state of two-level systems involving only levels 1 and 3.
[12]The idealizations and approximations used may limit the time during which the present theory is valid.

Free-Electron Laser with a Strong Axial Magnetic Field

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A small-signal theory is given for gain in a free-electron laser comprising a cold relativistic electron beam in a helical periodic transverse, and a strong uniform axial, magnetic field. Exact finite-amplitude, steady-state helical orbits are included. If perturbed, these orbits oscillate about equilibrium, so that substantial gain enhancement can occur if the electromagnetic perturbations resonate with these oscillations. This gain enhancement need not be at the cost of frequency upshift.

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Intensive activity is underway to exploit the gain properties of a relativistic electron beam undulating in a periodic transverse magnetic field. Such free-electron laser (FEL) configurations have provided oscillation at 3.4 (Ref. 1) and 400 μm,[2] and amplification at 10.6 μm.[3] Theory has advanced apace,[4] and elaborate schemes have been proposed for obtaining high FEL efficiency.[5] A factor which limits the practical application of this interaction at wavelengths shorter than perhaps a few microns is the rapid decrease in small-signal gain G_0 as the electron energy increases. This is shown explicitly in the well-known result[6] for G_0 in the single-particle limit (i.e., when collective effects are negligible)

$$G_0 = (\omega_p \xi / k_0 c)^2 (k_0 L / 2\gamma)^3 F'(\theta). \qquad (1)$$

Here ω_p and γ are the beam plasma frequency $Ne^2/m\epsilon_0$ and normalized energy W/mc^2, k_0 and ξ are the helical transverse magnetic field wave number $2\pi/l$ and normalized strength $eB_\perp/mck_0$, L is the interaction length, and $F'(\theta) = (d/d\theta)(\sin\theta/\theta)^2$ is the line-shape factor, with $\theta = [k_0 v_{30} - \omega(1 - v_{30}/c)](L/2c)$, where v_{30} is the unperturbed electron axial velocity. The peak gain occurs at $\theta = 1.3$, where $F'(\theta) = 0.54$. For example, with γ

