Resonance in an Oscillator with Two Nearby Frequencies: The Three-Level System

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The response to a monochromatic field of a three-level system, in which the (1, 2) interval is relatively close to the (2, 3) interval and the associated dipole matrix elements are arbitrary, is examined. It is found that, for weak pumping, proper detuning from exact two-photon resonance toward the more strongly coupled frequency produces a spin-$\frac{1}{2}$-type Rabi oscillation between levels 1 and 3, while proper detuning at strong pumping in the opposite direction produces a spin-1-type oscillation.

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The study of the behavior of various types of oscillators driven by monochromatic forces —essentially, the subject of resonance—is of fundamental interest. The response to a monochromatic force (MF) of a harmonic oscillator, which has a single natural frequency, is the simplest illustration of resonance. The response to a MF of an anharmonic oscillator, which has a continuous range of frequencies, classically, and a discrete range, quantum mechanically, is a more complicated problem, and has been the subject of a rich literature, both classical[1] and quantum mechanical,[2-4] motivated, recently, by efforts to produce selective molecular excitation by a laser field for purposes of isotope separation. Among the quantum mechanical studies, much attention has been given to oscillators with a finite number of levels.[5] The simplest and most fundamental of these is, of course, the two-level system, the response of which to a MF is well known and furnishes the basis for the understanding of many types of resonant phenomena. The next simplest, expectedly, is the three-level system, which has also received considerable attention, both exclusively,[6] and as a special case of a more general treatment of n-level systems.[3,5] In an analysis of the absorption of energy by this system, the interesting fact was discovered[3] that, for a weak MF, properly detuned from exact two-photon resonance in the direction of the more strongly coupled frequency, the three-level system behaves, approximately, like a two-level —or spin-$\frac{1}{2}$—system involving only levels 1 and 3; the occupation probability executes, approximately, a slow periodic (Rabi) oscillation between these two levels, while the occupation probability of level 2 oscillates rapidly with very small amplitude. It is the purpose of the present discussion to show that there exists another *type* of approximately periodic oscillation of the occupation probabilities, for a strong MF and for appropriate detuning in the direction of the *more weakly* coupled frequency, in which all three levels play an equal role, a type of oscillation that resembles the Rabi oscillation of a spin-1 system.[7]

Consider an atomic system of three levels, more or less evenly spaced, with energies $\hbar\omega_1$, $\hbar\omega_2$, $\hbar\omega_3$, in ascending order, and with only two nonvanishing dipole matrix elements, $\vec{\mu}_{12}$ and $\vec{\mu}_{23}$, corresponding to the frequencies ω_{12} and ω_{23}, where $\omega_{ij} \equiv |\omega_i - \omega_j|$. If the field acting on the system is given by $\vec{E} = 2\vec{E}_0 \cos\omega t$, the atomic Hamiltonian is specified by

$$\mathcal{H} = \sum \hbar\omega_i a_i^\dagger a_i + 2\hbar(\gamma_{12} a_1 a_2^\dagger + \gamma_{23} a_2 a_3^\dagger + \text{H.c.}) \cos\omega t,$$

where $\gamma_{ij} = -\vec{\mu}_{ij}\cdot\vec{E}_0/\hbar$. The notation has been described in detail previously.[8] Briefly, the a's and $a^\dagger$'s are boson annihilation and creation operators (with $[a_i, a_j^\dagger] = \delta_{ij}$), which describe a number of atoms behaving cooperatively. They can also be interpreted in the present calculation, if normalized by $\sum a_i^\dagger a_i = 1$, as single-atom energy-state amplitudes when expectation values are taken.

It is useful to introduce the notation $\Delta = \frac{1}{2}(\omega_{12} - \omega_{23})$, $\delta = \frac{1}{2}(\omega_{12} + \omega_{23}) - \omega$. The fact that our interest lies in a resonant phenomenon, and that the two atomic frequencies are relatively close, is indicated by the inequalities $\delta \ll \omega$, $\Delta \ll \omega$, respectively. These inequalities permit the use of the familiar rotating-wave approximation. With its use, and with the change of variables

$$a_1 = z_1 \exp[-i(\omega_1 + \delta)t], \quad a_2 = z_2 \exp[-i(\omega_2 - \Delta)t],$$
$$a_3 = z_3 \exp[-i(\omega_2 - \delta)t],$$

the equations of motion can be written

$$\dot{z}_1 - i\delta z_1 = -i\gamma_{12} z_2,$$
$$\dot{z}_2 + i\Delta z_2 = -i\gamma_{23} z_3 - i\gamma_{12} z_1,$$
$$\dot{z}_3 + i\delta z_3 = -i\gamma_{23} z_2.$$

Note that $\langle a_i^\dagger a_i\rangle = \langle z_i^\dagger z_i\rangle$, and that this quantity describes the probability of finding the atomic system in the ith level (when $\sum z_i^\dagger z_i$—a constant of motion—is set equal to unity). Since no interaction with other quantum mechanical systems is presently involved, we can treat the z_i's as c-numbers and ignore the expectation-value brackets.

We consider the case $z_1(0)=1$, $z_2(0)=z_3(0)=0$, and investigate $|z_2(t)|^2$ and $|z_3(t)|^2$. By use of the Laplace transformation, given by $\mathcal{L}\{z_i(t)\} = \int_0^\infty dt\, z_i(t)\exp(-st)$, the equations of motion can be solved routinely. The formal result is $z_i(t) = \mathcal{L}^{-1}\{N_i(s)/D(s)\}$, where $\mathcal{L}^{-1}$ indicates the inverse Laplace transformation, $N_2 = -i\gamma_{12}(s+i\delta)$, $N_3 = -\gamma_{12}\gamma_{23}$, and

$$D = s^3 + i\Delta s^2 + (\delta^2+\gamma^2)s + i\Delta\delta^2 + i\delta\alpha\gamma^2,$$

with $\gamma^2 \equiv \gamma_{12}^2 + \gamma_{23}^2$, and $\alpha \equiv (\gamma_{12}^2 - \gamma_{23}^2)/\gamma^2$. The three roots s_1, s_2, and s_3 of the cubic polynomial $D(s)$ determine the z_i's according to the formula

$$\mathcal{L}^{-1}\{(bs+c)/D\} = -W^{-1}\sum(bs_i+c)(s_j-s_k)\exp(s_i t),$$

the summation being over the three cyclic permutations of $i\neq j\neq k$, with $W=(s_1-s_2)(s_2-s_3)(s_3-s_1)$. The general expression for the roots in terms of the four parameters Δ, δ, γ^2, and α is too complicated to give physically transparent results. The present discussion will be restricted to certain ranges of the parameters that are both interesting, physically, and yield simpler exact or approximate expressions.

The simplest case is that of $\delta=0$, which may be regarded as the case of *exact two-photon resonance*, since the pump frequency is the mean of the two oscillator frequencies, and complete population inversion ($|z_3|^2=1$) corresponds to the absorption of two photons from the driving field. In this case $s_1=0$, $s_{2,3} = -\frac{1}{2}i\Delta \pm iM$, where $M\equiv(\gamma^2+\frac{1}{4}\Delta^2)^{1/2}$, and the occupation probabilities are given by[6]

$$|z_2|^2 = \tfrac{1}{2}\gamma^2(1+\alpha)M^{-2}\sin^2 Mt$$

$$|z_3|^2 = \tfrac{1}{4}(1-\alpha^2)[1+\cos^2 Mt - 2\cos\tfrac{1}{2}\Delta t\cos Mt - (\Delta/M)\sin\tfrac{1}{2}\Delta t\sin Mt + (\Delta^2/4M^2)\sin^2 Mt].$$

The familiar case of driven two-level system is described by setting $\gamma_{23}=0$ (or $\alpha=1$), a procedure that exhibits $2|\gamma_{12}|$ as the Rabi frequency associated with the two lower levels in the absence of the third. Analogously, $2|\gamma_{23}|$ may be regarded as the Rabi frequency associated with the upper pair of levels. Another simple exact solution is obtained for $\delta_0 = -\alpha\gamma^2/\Delta$, since this value also yields $s_1=0$. Inspection of D shows that the roots and $|z_3|^2$, for this case, can be obtained from the resonance case merely by the replacement of γ^2 by $\gamma^2+\delta_0^2$.

For other values of δ, none of the roots of D vanish, and we consider two ranges of the parameters for which at least one of the roots is small, so that perturbation theory can be used. These ranges are $\gamma^2 \ll \Delta^2$ and $\gamma^2 \gg \Delta^2$. Since γ is proportional to E_0, the first range may be considered that of weak pumping (with both Rabi frequencies small compared to the difference between the two atomic frequencies) and the second range that of strong pumping. In both ranges we look at the solution in the neighborhood of resonance, and consider $\delta^2 \ll \gamma^2$. For weak pumping, one obtains

$$|z_2|^2 = O(\gamma_{12}^2/\Delta^2),$$

$$|z_3|^2 = \gamma^4(1-\alpha^2)\Gamma^{-4}[\sin^2\tfrac{1}{2}(\Gamma^2/\Delta)t + O(\gamma^2/\Delta^2)],$$

where $\Gamma^4 = \gamma^4 + 4\Delta^2\delta^2 + 4\alpha\delta\gamma^2\Delta$, and quantities of order γ^2/Δ^2 are not written explicitly since they are irrelevant to the present argument. (It can be shown that $|z_2|^2$ does not differ qualitatively from that for weak pumping at resonance.) For strong pumping, to first order in Δ/γ and δ/γ, one obtains

$$|z_2|^2 \approx \tfrac{1}{2}(1+\alpha)[\sin^2\gamma t - 2(\delta/\gamma)\sin\gamma\beta t\sin\gamma t],\quad |z_3|^2 \approx \tfrac{1}{4}(1-\alpha^2)[1+\cos^2\gamma t - 2\cos\gamma\beta t\cos\gamma t - 2\beta\sin\gamma\beta t\sin\gamma t],$$

where $\beta \equiv (\Delta - 3\alpha\delta)/2\gamma$. (Setting $\alpha=\Delta=0$ yields the result for a spin-1 system.)

At resonance ($\delta=0$), the exact solution shows that $|z_3(t)|^2$ oscillates nonsinusoidally and nonperiodically, in general, and has local maxima with a range up to $1-\alpha^2$. The picture simplifies considerably for weak pumping ($\gamma^2/\Delta^2 \ll 1$) and for strong pumping ($\gamma^2/\Delta^2 \gg 1$). For weak pumping, the solution at resonance becomes, in lowest order,

$$|z_2|^2 \approx 2(\gamma^2/\Delta^2)(1+\alpha)\sin^2\tfrac{1}{2}\Delta t,$$

$$|z_3|^2 \approx (1-\alpha^2)\sin^2\tfrac{1}{2}(\gamma/\Delta)\gamma t.$$

For strong pumping, the solution at resonance becomes, in lowest order

$$|z_2|^2 \approx \tfrac{1}{2}(1+\alpha)\sin^2\gamma t,$$

$$|z_3|^2 \approx \tfrac{1}{4}(1-\alpha^2)(1+\cos^2\gamma t - 2\cos\tfrac{1}{2}\Delta t\,\cos\gamma t).$$

Here, the oscillation of $|z_3|^2$ is 50% modulated with frequency Δ. We note that the largest maximum of $|z_3|^2$ is $1-\alpha^2$.

Consider, now, the significance of the results for off-resonant pumping ($\delta \neq 0$). In the weak-pumping case, in lowest order, only $|z_3|^2$ and $|z_1|^2$ (where $|z_1|^2 \approx 1 - |z_3|^2$) are nonnegligible. For detuning given by $\delta = -\alpha\gamma^2/2\Delta$, the oscillation amplitude is maximized, so that

$$|z_3|^2 \approx \sin^2\tfrac{1}{2}(\gamma/\Delta)(1-\alpha^2)^{1/2}\gamma t.$$

This describes the Rabi oscillation of a two-level system driven on (the two-level) resonance, with complete population inversion. The effective (two-level) Rabi frequency is an order of magnitude (γ/Δ) smaller than the individual Rabi frequencies $2|\gamma_{ij}|$, and two orders of magnitude smaller than that of the small rapid oscillations of $|z_2|^2$. The fact that a weak field properly detuned from exact two-photon resonance can produce complete population inversion and spin-$\frac{1}{2}$-type periodic behavior in a three-level system was first deduced by Larsen and Bloembergen,[3] and has reappeared in other work on multilevel systems.[5] One can regard this phenomenon as the spin-$\frac{1}{2}$-type resonance of a three-level system. Inspection shows that the detuning brings the MF frequency closer to the frequency that couples *more strongly* to the field (with the larger γ_{ij}^2).

In the strong pumping case ($\gamma^2/\Delta^2 \gg 1$), we can see, by comparing the lowest-order terms, that the modulation frequency Δ of the $|z_3|^2$ oscillations in the resonant case is replaced by $\Delta - 3\alpha\delta$ in the off-resonant case. The essential effect, therefore, of tuning the field off resonance is the variation of this modulation frequency. By the choice $\delta = \Delta/3\alpha$, the modulation is eliminated entirely,[9] with all the maxima having the same (largest) value. The result is a periodic solution, in lowest order, given by

$$|z_2|^2 \approx \tfrac{1}{2}(1+\alpha)\sin^2\gamma t,$$

$$|z_3|^2 \approx \tfrac{1}{4}(1-\alpha^2)(1-\cos\gamma t)^2.$$

This is the only periodic solution for the $|z_i|^2$'s in the case of strong pumping. It resembles the Rabi oscillation of a spin-1 system driven near resonance, the only difference being the α terms. The energy oscillates with frequency γ, and both upper levels participate equivalently, in the sense that the population reaching the upper level may be regarded as going through the middle level in one cycle (of $|z_2|^2$).[10] The detuning δ necessary to produce this resonance brings the MF frequency closer to the *more weakly* coupled frequency, and can be interpreted as compensation for the weaker coupling. It is not unreasonable to suppose that such compensation exists also for higher multilevel systems. Finally, one should note that the strong-pumping case is the only case in which substantial two-photon excitation can occur in the presence of significant relaxation (absent in the present idealized model).

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[1]See, for instance, N. Minorsky, *Nonlinear Oscillations* (Van Nostrand, New York, 1962).

[2]L. M. Narducci *et al.*, Phys. Rev. A 16, 247 (1977).

[3]D. M. Larsen and N. Bloembergen, Opt. Commun. 17, 254 (1976).

[4]Y. Ben-Aryeh, Phys. Lett. 67A, 363 (1978), and references therein.

[5]J. H. Eberly *et al.*, Phys. Rev. A 16, 2038 (1977), and references therein; R. J. Cook and B. W. Shore, Phys. Rev. A 20, 539 (1979), and 20, 1958 (1979); B. W. Shore, Phys. Rev. A 24, 1413 (1981). In some of these references, independent driving fields, with generally different amplitudes and frequencies, act on the separate transition, respectively.

[6]R. G. Brewer and E. L. Hahn, Phys. Rev. A 11, 1641 (1975); D. Grischowsky *et al.*, Phys. Rev. A 12, 2514 (1975); F. T. Hioe and J. H. Eberly, Phys. Rev. A 25, 2168 (1982).

[7]A "spin-1 system" refers to an angular momentum oscillator of quantum number 1.

[8]I. R. Senitzky, Phys. Rev. A 10, 1868 (1974), and 15, 284 (1977). For simplicity, $\vec{\mu}_{ij}$ is taken to be real.

[9]It is assumed, here, that $\Delta/3\alpha$ is a small quantity of first order. Otherwise, the modulation cannot be eliminated by small detuning from resonance.

[10]Spin-1 type Rabi oscillation in a three-level system is a familiar type of behavior in the case where two independent driving fields act respectively (and exclusively) on the two transitions, provided resonance conditions are met and the two Rabi frequencies are equal. Such oscillation is discussed—as well as illustrated graphically—by M. Sargent III and P. Horowitz, Phys. Rev. A 13, 1962 (1976), by B. W. Shore and J. Ackerhalt, Phys. Rev. A 15, 1640 (1977), and by Eberly *et al.*, Ref. 5. When independent driving fields are available for the two transitions, the ratio of the Rabi frequencies can be controlled independently of the atom-

ic parameters by varying the field strengths; in fact, if the driving-field frequencies are resonant, respectively, with the transition frequencies, and the two Rabi frequencies are *equal,* the mathematical problem becomes similar to that of a spin-1 system driven on resonance. It should be noted, however, that such a phenomenon is different from the one considered presently. In the present instance of a single (monochromatic) driving field, the ratio of the two Rabi frequencies is determined solely by the atomic parameters; if the two transition frequencies are different, spin-1-type Rabi oscillation can be achieved only if the two Rabi frequencies are *unequal,* since the frequency shift from two-photon resonance required to produce this oscillation depends inversely on the parameter α, which vanishes for equal Rabi frequencies.

Rayleigh-Taylor Instabilities in Laser-Accelerated Targets

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Numerical studies of the ablation-driven Rayleigh-Taylor instability in laser-accelerated targets show growth rates typically within a factor of 2 of the classical growth rate. The appearance of the "Kelvin-Helmholtz" instability depends on the form of the initial perturbation and also on the laser irradiance. Perturbations of the target surface and laser irradiation are simulated and compared.

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The achievement of inertially confined thermonuclear fusion in laser driven pellets requires that a hollow shell be symmetrically imploded to less than one tenth of its initial radius in order to generate the high densities needed for significant thermonuclear burn.[1] The use of a hollow rather than solid pellet reduces the peak power requirement from the laser which decreases as the ratio of shell radius to thickness, $\alpha = R/\Delta R$, is increased. Unfortunately the hollow shell targets are hydrodynamically unstable in the ablation region where the pressure and density gradients are of opposite sign, i.e., $\nabla p \cdot \nabla \rho < 0$. The instability in this region is similar to the classical Rayleigh-Taylor (RT) instability of two incompressible fluids[2] but is complicated by the finite density and temperature scale lengths, heat conduction, compressibility, and flow of the ablating material. Various analytic approximations have been made to estimate the growth rates of the instability,[3,4] and numerical simulation with use of both Eulerian and Lagrangian formulations have also been employed.[5-7]

The simulation data consistently show growth rates γ reduced by a factor of 2 or 3 below the classical growth rate for an Attwood number $\gamma = (ka)^{1/2}$ of unity, where k is the wave number and a the effective acceleration.

There is a qualitative difference between the results of McCrory *et al.*[6] and those of Emery, Gardner, and Boris[7] regarding the appearance of a Kelvin-Helmholtz (KH) type of instability as evidenced by a broadening of the tips of the RT "spikes" as they fall into the less dense medium. These differences have at times been attributed to the numerical differences of the Eulerian and Lagrangian formulations, and particularly to a supposed "stiffness" of the triangular Lagrangian mesh induced by the von Neumann artificial viscosity used in all Lagrangian codes. We report here the results of simulations performed with an Eulerian code (which does not use an artificial viscosity) which show that the appearance of the "KH" features is dependent on the initial conditions of the problem and cast some doubt as to whether it is indeed a KH instability. Our results agree well with both Emery, Gardner, and Boris and McCrory *et al.* for the rather dissimilar cases that they considered.

The simulations are performed with an Eulerian formulation in cylindrical (r, z) geometry. The code includes laser absorption by inverse bremsstrahlung, and electron and ion energy transport by flux-limited thermal conduction and fluid mo-

