Atomic excitation by a multimode symmetric laser

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The excitation of an atom by a near-resonant-symmetric mode-locked multimode laser is solved in the two-state rotating-wave approximation. An analytic expression of an unusual new form of resonance is found and a simple experiment which could be used to observe the effect is suggested.

I. INTRODUCTION

Most calculations of atomic processes in laser fields start from an ideal laser whose amplitude is given in dipole approximation by

$$\mathbf{E}(t)=\mathbf{E}_0\cos(\omega t+\theta) \tag{1.1}$$

or its circularly polarized analog where $\mathbf{E}_0$, ω, and θ are constants in time. Most experiments are performed with multimode lasers with an amplitude which can be approximated by

$$\mathbf{E}(t)=\sum_i \mathbf{E}_i(t)\cos[\omega_i t+\theta_i(t)]\ , \tag{1.2}$$

where the sum runs over the modes, and both the amplitudes and phases are functions of time. If their variation is slow on a time scale set by the atomic process under study then the result based upon (1.1) can be ensemble averaged[1] to get the result relevant to (1.2). If the variation of the parameters in (1.2) is too fast for this procedure to be valid then a more general,[2] and more complex, procedure can be used to describe the atomic process. This technique relies upon an average over the many modes of the laser at the outset of the calculation and may obscure some of the physics in the process. The treatment of the chaotic laser or the phase diffusion laser in this way requires an average over the random phases in (1.2) which suppresses the possible effects of mode beating which occur with a mode-locked laser.

We shall investigate some of these effects analytically by the consideration of a very special, but realistic, symmetric three-mode laser,

$$\mathbf{E}=[\mathbf{E}_0+\mathbf{E}_1\cos(\eta t)]\cos(\omega t+\theta) \tag{1.3}$$

($\eta \ll w$) which is slightly detuned from a transition between two atomic states u_0,u_1 with energies W_0,W_1. The detuning is given by

$$\Delta\omega=\omega-(W_1-W_0) \tag{1.4}$$

which is assumed to be small compared to η. The couplings between the two states induced by (1.3) are

$$\Lambda_i=\mathbf{d}_{01}\cdot\mathbf{E}_i,\ \ i=0,1 \tag{1.5}$$

where $\mathbf{d}_{01}$ is the dipole moment connecting the states.

In Sec. II the generalization of the two-state rotating-wave approximation is performed to obtain the recoupled atomic states first for the case in which the parameters of (1.3) are constants and then for the case in which the laser is adiabatically turned on and off. We find a new kind of resonance in the excitation process either as a function of ω, or as a function of the amplitude of the central mode. Finally, we suggest and analyze an experiment which could be used to observe the effect. The results can be generalized to a multimode symmetric mode-locked laser of the form [generalizing (1.3)]

$$\mathbf{E}=\left[\mathbf{E}_0+\mathbf{E}_1\cos(\eta t)+\sum_{j=2}^{N}\mathbf{E}_j\cos(j\eta t+\theta_j)\right]\cos(\omega t+\theta) \tag{1.6}$$

with all the $\theta_j=0$. This is described briefly for $N=2$ and is easily generalized to higher N. Finally, the results are discussed in Sec. III.

There is a long history of consideration of the effect of multimode lasers on atoms: An atom moving in a single-mode standing-wave laser field will Doppler shift the two equivalent traveling-wave fields differently so that in the atom's rest frame the laser will appear to be bimodal.[3] Another example is the absorption of the beats of two different lasers by the nonlinearities of the atomic interaction.[4] The response of an atom to a modulated laser field has been considered in the context of phase-shift measurements of lifetimes.[5] Fluorescence in weakly modulated laser fields has also been considered.[6] The work closest to this one, of which I am aware, by Agarwal and Nayak,[7] concerns the calculation of the nonlinear susceptibility of a two-level atom in a bimodal field. The two modes were described by a phase-diffusion model so an average over the phases was performed. Nevertheless the numerical results showed that resonances in the absorption spectrum as a function of (the analog of) η still survive. The work presented here appears to be the first analytic description of the resonances in the response of a two-level atom to a multimode laser.

II. TWO-STATE ROTATING-WAVE APPROXIMATION

The laser is almost resonant with the two atomic states and weak enough so that the dynamic Stark shifts may be neglected. Then a good approximation for the atomic states in the laser field (1.3) is ($\hbar=1$)

$$\psi=u_0e^{-i(W_0-\Delta\omega/2)t}\alpha+u_1e^{-i(W_1+\Delta\omega/2)t}\beta\ , \tag{2.1}$$

where the parameters satisfy

$$i\dot{\alpha}=\tfrac{1}{2}\Lambda\beta+\tfrac{1}{2}\Delta\omega\,\alpha\ ,$$
$$i\dot{\beta}=\tfrac{1}{2}\Lambda^*\alpha-\tfrac{1}{2}\Delta\omega\,\beta\ , \tag{2.2}$$

where

$$\Lambda=\mathbf{d}_{01}\cdot(\mathbf{E}_0+\mathbf{E}_1\cos\eta t)e^{i\theta}\ . \tag{2.3}$$

The phases of the states u_0 and u_1 may be adjusted to make Λ real so that this can be written

$$\Lambda(t)=\Lambda_0+\Lambda_1\cos(\eta t)=\Lambda^*(t)\ . \tag{2.4}$$

If we define a column matrix

$$A=\begin{bmatrix}\alpha\\ \beta\end{bmatrix} \tag{2.5}$$

then (2.2) can be written

$$i\dot{A}=\tfrac{1}{2}\{[\Lambda_0+\Lambda_1\cos(\eta t)]\sigma_x+\Delta\omega\,\sigma_z\}A\ , \tag{2.6}$$

where the σ_i are the usual 2×2 Pauli matrices. This equation can be rewritten as a single second-order linear differential equation with periodic coefficients. The machinery of Hill's determinant is then appropriate for the explicit solution. This leads to numerical calculations which are more general but not as illuminating as the analytic results obtained below. Therefore, the substitutions

$$A=e^{-(i/2)\phi\sigma_x}B,$$
$$\phi=\Lambda_0 t+\Lambda_1/\eta\sin(\eta t),\quad \tau=\Delta\omega\, t\ , \tag{2.7}$$

yield

$$i\frac{dB}{d\tau}=\tfrac{1}{2}(\sigma_z\cos\phi+\sigma_y\sin\phi)B\ , \tag{2.8}$$

where we can write

$$\cos\phi=\sum_{n=-\infty}^{\infty}\cos[(\Lambda_0+n\eta)\tau/\Delta\omega]\ , \tag{2.9a}$$

$$\sin\phi=\sum_{n=-\infty}^{\infty}\sin[(\Lambda_0+n\eta)\tau/\Delta\omega]\ . \tag{2.9b}$$

For general n the terms in the sum in (2.9) are rapidly varying functions of τ and will contribute terms of order $\Delta\omega/(\Lambda_0+n\eta)$ to B which is small. However, there is a particular integer $n=\nu$ where this *may* be significant. We define

$$\nu=-[\Lambda_0/\eta] \tag{2.10}$$

which is the integer closest to $-\Lambda_0/\eta$. That term may contribute significantly if

$$\Omega=\Lambda_0+\nu\eta \tag{2.11}$$

is much smaller than η and if $J_\nu(\Lambda_1/\eta)$ is not small. We retain only this term in (2.8). Then

$$i\frac{dB}{d\tau}=\tfrac{1}{2}J_\nu(\Lambda_1/\eta)\{\cos[(\Omega/\Delta\omega)\tau]\sigma_z+\sin[(\Omega/\Delta\omega)\tau]\sigma_y\}B$$
$$=\tfrac{1}{2}J_\nu(\Lambda_1/\eta)e^{i(\Omega/2\Delta\omega)\tau\sigma_x}\sigma_z e^{-i(\Omega/2\Delta\omega)\tau\sigma_x}B\ . \tag{2.12}$$

It is easily shown that the solution to (2.12) is

$$B(t)=e^{(i/2)\Omega t\sigma_x}e^{-(i/2)(\Delta\omega J_\nu\sigma_z+\Omega\sigma_x)t}B(0)\ , \tag{2.13}$$

which results in

$$A(t)=e^{(i/2)[\Omega t-\phi(t)]\sigma_x}\times e^{-(i/2)(\Delta\omega J_\nu\sigma_z+\Omega\sigma_x)\tau}A(0)[1+O(\Delta\omega/\eta)]\ . \tag{2.14}$$

One can carry through the algebra implied by the exponential operators with the aid of the definition

$$\epsilon=[(\Delta\omega)^2J_\nu^2(\Lambda_1/\eta)+\Omega^2]^{1/2}\ , \tag{2.15}$$

and substitute back into (2.1) with the result

$$\psi=\alpha(0)\psi_0+\beta(0)\psi_1\ , \tag{2.16}$$

where

$$\psi_0=x_1(t)u_0e^{-i(W_0-\Delta\omega/2)t}-x_2^*(t)e^{-i(W_1+\Delta\omega/2)t}u_1\ ,$$
$$\psi_1=x_2(t)u_0e^{-i(W_0-\Delta\omega/2)t}+x_1^*(t)e^{-i(W_1+\Delta\omega/2)t}u_1\ , \tag{2.17}$$

and where

$$2x_1=(1-\Omega/\epsilon)\cos\zeta_+ +(1+\Omega/\epsilon)\cos\zeta_- +i\frac{\Delta\omega J_\nu}{\epsilon}(\sin\zeta_+-\sin\zeta_-)\ ,$$
$$2x_2=\frac{\Delta\omega\, J_\nu}{\epsilon}(\cos\zeta_+-\cos\zeta_-)-i(1-\Omega/\epsilon)\sin\zeta_+-i(1+\Omega/\epsilon)\sin\zeta_-\ , \tag{2.18}$$

with

$$\zeta_\pm(t)=\tfrac{1}{2}[\phi(t)-(\Omega\pm\epsilon)t]\ . \tag{2.19}$$

The states ψ_0 and ψ_1 are each normalized to unity and are orthogonal to each other. For $t=0$, $\psi_0=u_0$ and $\psi_1=u_1$.

We may ask for P_1, the probability of finding u_1 in ψ_0 at some later time:

$$P_1(t)=|x_2(t)|^2$$
$$=\frac{1}{2}\left[1-\frac{\Omega}{2\epsilon}\left[1+\frac{\Omega}{\epsilon}\right]\cos(2\zeta_-)+\frac{\Omega}{2\epsilon}\left[1-\frac{\Omega}{\epsilon}\right]\cos(2\zeta_+)-\frac{2(\Delta\omega)^2J_\nu^2}{\epsilon^2}\cos(\zeta_++\zeta_-)\right]. \quad (2.20)$$

This can be rewritten by using a relation similar to (2.9) with the result

$$P_1(t)=\frac{1}{2}\left[1-\frac{\Delta\omega^2J_\nu^2}{\epsilon^2}\sum_n J_n(\Lambda_1/\eta)\cos[(n-\nu)\eta t]-\frac{\Omega}{2\epsilon}\left[1+\frac{\Omega}{\epsilon}\right]\sum_n J_n(\Lambda_1/\eta)\cos[(n-\nu)\eta t+\epsilon t]-\frac{\Omega}{2\epsilon}\left[1-\frac{\Omega}{\epsilon}\right]\sum_n J_\nu(\Lambda_1/\eta)\cos[(n-\nu)\eta t-\epsilon t]\right]. \quad (2.21)$$

The time average of this is

$$\bar{P}_1=\frac{1}{2}\left[1-\frac{(\Delta\omega)^2J_\nu^3(\Lambda_1/\eta)}{\epsilon^2}\right], \quad (2.22)$$

which exhibits a resonance behavior at $\Delta\omega=0$ with a resonance width given by

$$\left[\frac{\Gamma}{2}\right]^2=\frac{\Omega^2}{J_\nu^2(\Lambda_1/\eta)}. \quad (2.23)$$

The sign of the contribution to the structure in P_1 depends upon the sign of J_ν. There is also a resonancelike structure as a function of E_0 (via Λ_0 in Ω). It is not a result of the dynamic Stark effect which can also induce resonances as a function of E_0.

This is a rather unusual form for a resonance structure but we can provide some qualitative understanding as follows. Suppose Λ_1 is very small, then we return to the usual problem of an ideal laser and a two-state rotating-wave approximation.[3] The result is a pair of "dressed" states separated in quasienergy by essentially $|\Lambda_0|$. A weak probe laser (Λ_1) can resonantly connect these states and there will be a rapid variation of the probability of this connection with the probe laser frequency (η) near $\eta=|\Lambda_0|$. Equation (2.22) may be thought of as the nonlinear generalization of this process.[8]

We now turn to the generalization of these results to the case of the adiabatic switching on and off of the laser amplitude. Then Λ_0 and Λ_1 become slowly varying functions of time which vanish for $t\to\pm\infty$. This can be incorporated in the results above by the replacement $\Lambda_i\to\Lambda_i(t)$ and $\Lambda_i t\to\int_{-\infty}^{t}dt'\Lambda_i(t')$ in all the above results. This implies that $\epsilon t\to\int_{-\infty}^{t}dt'[\epsilon(t')-\epsilon(\infty)]+\epsilon(\infty)t$. The errors incurred are of order T^{-1} where T is the time scale of adiabatic variation of $\Lambda_i(t)$. There is an additional time dependence entering through ν which depends upon Λ_0, (2.10), in a discontinuous fashion. This will introduce corrections to the adiabatic solution which are proportional to $\dot{\nu}$ which is not small at the points of discontinuity of ν. The difficulty can be avoided by stipulating that ν is a constant. This can be achieved by limiting $\nu\le\frac{1}{2}$ or by making $\eta\sim\Lambda_0(t)$ so that ν is a constant. This adiabatic variation of η [see (1.3)] is then handled by the replacement $\eta t\to\int_{-\infty}^{t}dt'\eta(t')$ in all the equations above. (This mathematical device may in fact make the analogous experiment too difficult to perform.) Then, in summary, the results described above may be carried over to the adiabatic switching case with these replacements (with errors of order T^{-1}), the only change being that for $t=-\infty$, $\psi_0=u_0$ and $\psi_1=u_1$.

An experiment which can be used to investigate these phenomena can be performed by using a second weak probe laser [$E'\cos(\omega' t)$] to excite the transition from u_1 to another state u_2. Then the radiative decay of u_2 can be monitored as a function of the parameters of the system. The transition $u_1\to u_2$ can be treated by first-order perturbation theory (Fermi's golden rule) as a transition from the state dressed by the first laser, ψ_0, to u_2. The main contribution to the result comes from the u_1 component of ψ_0 which after some manipulation, yields a matrix element

$$(u_2e^{-iW_2t},\mathbf{d}\cdot\mathbf{E}'e^{-i\omega't}\psi_0)=-\mathbf{d}_{21}\cdot\mathbf{E}'\frac{\pi}{2}\sum_n\Bigg\{\delta(W_{21}-\omega'-\tfrac{1}{2}\Delta\omega+\tfrac{1}{2}\eta(n-\nu)-\tfrac{1}{2}\epsilon)\times\left[\left[\frac{\Delta\omega J_\nu(\Lambda_1/\eta)}{\epsilon}-1+\frac{\Omega}{\epsilon}\right]J_n(\Lambda_1/2\eta)-\left[\frac{\Delta\omega J_\nu(\Lambda_1/\eta)}{\epsilon}-1-\frac{\Omega}{\epsilon}\right]J_{2\nu-n}(\Lambda_1/2\eta)\right]$$
$$+\delta(W_{21}-\omega'-\tfrac{1}{2}\Delta\omega-\tfrac{1}{2}\eta(n-\nu)+\tfrac{1}{2}\epsilon)\times\left[\left[\frac{\Delta\omega J_\nu(\Delta_1/\eta)}{\epsilon}+1-\frac{\Omega}{\epsilon}\right]J_n(\Lambda_1/2\eta)-\left[\frac{\Delta\omega J_\nu(\Lambda_1/\eta)}{\epsilon}+1+\frac{\Omega}{\epsilon}\right]J_{2\nu-n}(\Lambda_1/2\eta)\right]\Bigg\}. \quad (2.24)$$

Barring accidental degeneracies (which lie outside the scope of this calculation) each of the energy delta functions yields a contribution which adds incoherently with all the others. For example, the weak laser can be tuned so that it selects only the $n=\nu$ terms. Then the transition rate to u_2 is proportional to

$$|\mathbf{d}_{21}\cdot\mathbf{E}'|^2\frac{\Omega^2}{\epsilon^2}J_\nu^2(\Lambda_1/2\eta)[\delta(W_{21}-\omega'-\tfrac{1}{2}\Delta\omega-\tfrac{1}{2}\epsilon)+\delta(W_{21}-\omega'-\tfrac{1}{2}\Delta\omega+\tfrac{1}{2}\epsilon)]\ . \tag{2.25}$$

Other values of n yield somewhat more complex forms but they all have the ϵ^{-2} dependence which yields resonance at $\Delta\omega=0$ with width

$$\Gamma/2=|\Omega/J_\nu(\Lambda_1/\eta)|\ , \tag{2.26}$$

as in (2.22). This may indeed be a measurable result.

Incoherent effects such as collisions and fluorescence can be inserted phenomenologically but this is unnecessary if the transitions are saturated by the laser and if the experiment is performed with a short-pulsed laser. The deductive investigation of fluorescence is a nontrivial problem.

We now turn to the generalization to more modes with a laser of the form (1.6) with $N=2$. Then the new ϕ in (2.8) is

$$\phi=\Lambda_0 t+\frac{\Lambda_1}{\eta}\sin(\eta t)+\frac{\Lambda_2}{2\eta}\sin(2\eta t+\theta_2)\ , \tag{2.27}$$

and (2.9) becomes

$$\cos\phi=\sum_{n_1,n_2}J_{n_1}(\Lambda_1/\eta)J_{n_2}(\Lambda_2/2\eta)\times\cos[(\Lambda_0+n_1\eta+2n_2\eta)t+n_2\theta_2]\ , \tag{2.28a}$$

$$\sin\phi=\sum_{n_1,n_2}J_{n_1}(\Lambda_1/\eta)J_{n_2}(\Lambda_2/2\eta)\times\sin[(\Lambda_0+n_1\eta+2n_2\eta)t+n_2\theta_2]\ , \tag{2.28b}$$

The argument leading to (2.11) is unchanged so we must now select the slowly varying terms arising from (2.28) given by

$$n_1+2n_2=\nu \tag{2.29}$$

so that the generalization of (2.12) becomes

$$i\frac{dB}{d\tau}=\tfrac{1}{2}\sum_{n_2}J_{\nu-2n_2}(\Lambda_1/\eta)J_{n_2}(\Lambda_2/2\eta)\times e^{+(i/2)(\Omega\tau/\Delta\omega+n_2\theta_2)\sigma_x}\sigma_z\times e^{-(i/2)(\Omega\tau/\Delta\omega+n_2\theta_2)}B\ . \tag{2.30}$$

The n_2 dependence in the exponentials in this equation precludes a simple solution for B. However, if the five-mode laser is specialized to $\theta_2=0$, then (2.30) becomes

$$i\frac{dB}{d\tau}=\tfrac{1}{2}\sum_{n_2}J_{\nu-2n_2}(\Lambda_1/\eta)J_{n_2}(\Lambda_2/2\eta)e^{i(\Omega/2\Delta\omega)\tau\sigma_x}\times\sigma_z e^{-i(\Omega/2\Delta\omega)\tau\sigma_x}B \tag{2.31}$$

which is a simple generalization of (2.12) with

$$J_\nu(\Lambda_1/\eta)\rightarrow\sum_{n_2}J_{\nu-2n_2}(\Lambda_1/\eta)J_{n_2}(\Lambda_2/2\eta)\ . \tag{2.32}$$

Then the subsequent equations of this section can be carried over with this simple modification. The solution for higher values of N in (1.6) can be obtained in a similar way. Equation (2.32) is an indication of the complexity of the nonlinear combination of the various modes in (1.6) to form the resonance in the excitation.

III. DISCUSSION

There are some interesting implications of the results obtained above. For example, with a single-mode laser the probability of finding the excited state when we start from the ground state is

$$P_1=\frac{1}{2}\left[1-\frac{(\Delta\omega)^2}{(\Delta\omega)^2+\Lambda^2}\right]\ . \tag{3.1}$$

The excitation is effective over a frequency band given roughly by $\Delta\omega\sim\pm\Lambda$ and Λ is interpreted as the "power broadening" of the states. For a broadband laser it is not at all clear as to what the power broadening should be. How much can (the central frequency of) the broadband laser be detuned and still give significant excitation? Equation (2.22) yields a result $|\Omega/J_\nu(\Lambda_1/\eta)|$ for the power broadening defined in this way. (Note that the constraint $|\Delta\omega/\eta|\ll1$ is built into the theory of the outset.) For a weak laser we obtain $\nu=0$, $\Lambda_1/\eta\ll1$ and

$$\tfrac{1}{2}\Gamma=\left|\frac{\Omega}{J_\nu}\right|\rightarrow|\Lambda_0|$$

so the single-mode laser result is recovered. [Note that $\Lambda_1/\eta\ll1$ is precluded for all but $\nu=0$ by the requirement given below (2.10).]

The theory of Sec. II is only valid when $\Omega\ll\eta$, otherwise the terms dropped in solving (2.8) are the same order

TABLE I. Width $|\Gamma/2\Lambda_0|$ (2.26) for various values of $X=|\Lambda_0/\eta|$ and Λ_0/Λ_1.

X	ν	$\Lambda_0/\Lambda_1=2$	$\Lambda_0/\Lambda_2=1$	$\Lambda_0/\Lambda_1=0.5$
2.2	−2	0.67	0.23	0.36
2.1	−2	0.38	0.13	0.15
1.9	−2	0.50	0.16	0.13
1.8	−2	1.17	0.36	0.25
1.2	−1	0.58	0.33	0.32
1.1	−1	0.34	0.19	0.16
0.9	−1	0.51	0.27	0.19
0.8	−1	1.28	0.68	0.44
0.2	0	1.00	1.01	1.04
0.1	0	1.00	1.00	1.01

of magnitude as those retained. Within this constraint we may investigate the width, (2.26), in units of Λ_0 for various values of Λ_0/Λ_1. This is shown in Table I for various values of $X = |\Lambda_0/\eta|$ and ν [Eq. (2.10)]. Integer values of X are omitted since $\Gamma=0$ and the result (2.25) is also zero for that case.

We see that $\Gamma/2\Lambda_0$ is essentially unity for a very broad-band laser ($X=\Lambda_0/\eta \ll 1$) which is the usual single-mode-laser result. As X is increased, and the side modes couple more strongly, the width tends to *decrease.*

These results have been derived on the basis of a mode-locked symmetric laser [(1.6) with $\theta_j=0$]. It is not at all clear how they would change when the symmetric and mode-locked restrictions are removed. We hope to investigate this in the future.

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