Time-dependent aspects of the Autler-Townes effect in a four-level system

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Time-dependent optical Autler-Townes spectra are calculated for a four-level system in which the ground state is a doublet with frequency separation much larger than any of the Rabi frequencies. The finite linewidths of the lasers are taken into account in the phase-diffusion model. Optical-pumping effects cause a transfer of population with a resulting significant change in the spectra with time. Numerically evaluated spectra are presented, and it is found that the nature of the asymmetry can change drastically with time.

I. INTRODUCTION

The theory of the optical Autler-Townes effect for a three-level system is reasonably tractable,[1] and a symmetric, doubly peaked spectrum is predicted when the saturating transition is exactly resonant. The simplest situation is where the experiments are performed on an atomic beam so that Doppler and collision effects are negligible; the predictions for this case have been recently confirmed in elegant experiments.[2] The case of Doppler-broadened three-level systems has also been considered.[3] Again, reasonably good agreement between theory and experiment is found.

In making the calculations, it is usually assumed that the atoms interact with the laser field sufficiently long for the system to reach a steady state. However, it is clear that if the experiments are performed with pulsed lasers of sufficiently short duration or with atomic beams with sufficiently small interaction volume, this condition may not be met. For example, with a typical laser beam diameter of 10^{-3} m and atomic lifetime of 10^{-7} s, the atom is in the laser beam for about ten atomic lifetimes. Moreover, experiments have been performed with laser beams of one-tenth of this diameter,[4] giving a transit time of the order of 1 excited-state lifetime. It is therefore of some interest to calculate the time-dependent spectra, and to estimate the illumination time necessary for the steady-state solution to be a good approximation.

One possibility is that asymmetry may be introduced if the steady-state conditions are not met, and in some of the experiments reported on three-level systems,[2] there is some suggestion that the spectra are not completely symmetric. However, recent calculations[5] relevant to sodium have shown that although the asymmetry of the Autler-Townes spectrum for a three-level system does vary with the interaction time, the effect is small.

A similar situation occurs in the related phenomenon of strong-field resonance fluorescence. Experiments performed on atomic beams of sodium prepared as a two-level system under resonant excitation (e.g., Ref. 6) indicate that under some circumstances slight asymmetries are observable. It has been suggested (e.g., Ref. 7) that these may be due to finite interaction times. Cohen-Tannoudji and Reynaud[8] have emphasized that steady-state conditions need not obtain in actual experiments. A detailed discussion of the time-dependent resonance fluorescence spectrum has been given by Eberly *et al.*,[9] and they find that the asymmetry of the spectrum varies markedly with time under some circumstances.

All the preceding discussion refers to three-level systems (or two-level systems, for resonance fluorescence). However, experiments have been performed on multilevel systems.[10,11] Picqué and Pinard,[10] for example, made measurements of the optical Autler-Townes spectrum of sodium without it being prepared as a three-level system, and found a doubly peaked asymmetric spectrum at exact resonance. The theory, taking into account the full hyperfine structure of sodium, but neglecting the laser linewidths and calculating the steady-state spectrum, has been given,[12] but it is found that the predicted asymmetry is opposite to that observed. Related experiments have been made by Hogan *et al.*,[11] who monitored the Autler-Townes splitting in sodium by observing the multiphoton ionization. The theory modeling sodium as a three- or four-

level system, has been given by Georges and Lambropoulos.[13] The duration of the laser pulses used in the experiments was about 700 nsec with a rise time of about 75 nsec; thus it was necessary to take into account the finite interaction time which was done by numerically solving the differential equations. It is also essential to take into account the finite laser linewidths; Georges and Lambropoulos pointed out that the asymmetry of the spectrum is reversed as the laser linewidth increases from zero to a value greater than the natural linewidth. Zoller, Dixit, and Lambropoulos[14] emphasized the importance of non-Lorentzian line shapes.

The theory of an N-level atomic system coupled to monochromatic lasers which are near resonance with the $N-1$ transitions has been discussed analytically and numerically by Eberly, Shore, Bialynicka-Birula, and Bialynicki-Birula.[15] These authors also considered an anharmonic ladder of states which is relevant to molecular interactions.

In this paper we consider the time dependence of the optical Autler-Townes spectra in a four-level atomic system illuminated with finite-bandwidth lasers. The ground state is assumed to a doublet with energy separation small compared with the energy of the laser photons but large compared with the Rabi frequencies of the principal transitions. (This may be taken to be a crude model for sodium, for example.) The situation is shown schematically in Fig. 1. It is clear that we have a very different situation here from the three-level system because of optical pumping; if the atom is prepared initially in one of its ground states, some population will be transferred to the other ground state by laser action and spontaneous emission, and this implies that the Autler-Townes spectrum will change drastically with time. Thus we would expect that finite transit times will be much more significant in a four-level model than in a three-level system. It is clear, from the work of Georges and Lambropoulos,[13] that finite laser linewidths must also be taken into account. Here we treat the laser fields in the phase-diffusion model and take the line shapes to be Lorentzian. It is assumed that the only other relaxation mechanism is spontaneous decay (no collisions, etc.). The transient response will depend upon the laser pulse shape in time but for simplicity we consider here only the case in which the pulse shape is a step function.

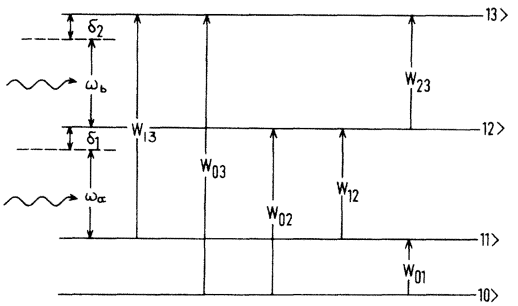

FIG. 1. Four-level model showing the "upward" transition rates.

In Sec. II we describe the model and method of solution, in Sec. III we introduce the probe-field approximation, and in Sec. IV we present the numerical results which show, for some choices of the parameters at any rate, that the asymmetry of the Autler-Townes spectra in a four-level system are marked time dependent.

II. THEORY

A. The model

We consider a four-level atomic system as shown in Fig. 1. The ground state is assumed to consist of two nearly degenerate states, denoted $|0\rangle$ and $|1\rangle$, with corresponding energies E_0 and E_1; the other two states $|2\rangle$ and $|3\rangle$ are excited states with energies $E_2 < E_3$. (For simplicity we assume a system of units in which $\hbar=1$, so that energy and frequency have the same dimensions.) An intense laser, A, is tuned to near resonance with the ground state $|1\rangle$ and the excited state $|2\rangle$: Its frequency is denoted w_a. A weak laser, B ("the probe laser") of frequency w_b is near resonant with the $|2\rangle \leftrightarrow |3\rangle$ transition.

The interaction between the atom and the fields is assumed to be of the usual dipole form

$$V=\sum_{\lambda=a,b}\sum_{i,j=0}^{3} i\left[\frac{2\pi\alpha\omega_\lambda}{\Omega}\right]^{1/2}(r_{ij}a_\lambda - r_{ij}^{*}a_\lambda^{\dagger}) , \qquad (1)$$

where Ω is the volume of the system, α the fine-structure constant, a_λ the annihilation operator for the λth laser, and r_{ij} the dipole matrix element for the atomic transition $|i\rangle \leftrightarrow |j\rangle$. For simplicity, we assume there is no direct (one-photon) transition between the ground states and level $|3\rangle$, or between the two components of the ground states, i.e., we assume

$$r_{03}=r_{13}=r_{01}=0 . \qquad (2)$$

There is still the possibility of indirect (two-photon) transitions between these states.

If we assume the two laser fields are monochromatic, with initially n_a photons in the A laser field and n_b photons in the B laser field, then within the rotating wave approximation only the composite laser/atom states

$$|0;n_a,n_b\rangle,|1;n_a,n_b\rangle,|2;n_a-1,n_b\rangle,|3;n_a-1,n_b-1\rangle \tag{3}$$

are connected by the interaction (1). (We have assumed that laser A causes no transitions between levels $|2\rangle$ and $|3\rangle$, and that laser B causes no transitions between levels $|0\rangle$, $|1\rangle$, and $|2\rangle$. The principal physical effect neglected by this assumption is the light shifts induced in the atomic levels by the opposing lasers; these can be made negligibly small by taking the resonant frequencies $\omega_a \sim E_2-E_1$ and $\omega_b \sim E_3-E_2$ to be sufficiently different.) It is clear that these states are unambiguously defined if we specify only the atomic part. Henceforth, for notational simplicity, we use $|0\rangle$, $|1\rangle$, $|2\rangle$, $|3\rangle$ to denote the composite states $|0;n_a,n_b\rangle$, $|1;n_a,n_b\rangle$, $|2;n_a-1,n_b\rangle$, and $|3;n_a-1,n_b-1\rangle$, respectively. We can take into account the finite linewidths of the lasers as indicated in Ref. 5. This assumes that the broadening mechanism is phase diffusion (e.g., Ref. 16), and that the laser line shape is Lorentzian:

$$J_\lambda(\omega)=\frac{J_\lambda \Delta_\lambda/\pi}{(\omega-\omega_\lambda)^2+\Delta_\lambda^2}, \quad \lambda=a,b \tag{4}$$

where J_λ is the total energy flux of the λth laser, ω_λ its center frequency, and Δ_λ its half-width. (In fact, the spectrum for a phase-diffusion-broadened laser falls off more rapidly than a Lorentzian in the wings.[16,14])

B. General solution for N-level system

As our method of solution has already been described in Refs. 5 and 17, we do not repeat the details here. However, we take the opportunity of presenting the general solution for an arbitrary N-level system, and then discuss the solution for our four-level model in this context.

The rate equations for the Laplace transforms $\tilde{P}_i(z)$ of the time-dependent probabilities $P_i(t)$ of being in the system state $|i\rangle$ are given, according to Ref. 17, by

$$(z+W_i)\tilde{P}_i-\sum_{j\neq i} W_{ji}\tilde{P}_j=P_i(0), \quad i=0,1,\ldots,N-1 \tag{5}$$

where W_i is the total rate of transitions out of level $|i\rangle$:

$$W_i=\sum_{j\neq i} W_{ij}\ , \tag{6}$$

and in Eqs. (5) and (6) we have suppressed the z dependence of the $\tilde{P}_i$ and the "generalized transition rates," W_{ij}, for brevity. Equation (5) represents a balance between the total rate of transitions *out* of level $|i\rangle$ (second term on the left-hand side) and the total rate of transitions *into* level $|i\rangle$ from other levels $|j\rangle$ (third term on left-hand side). The term $P_i(0)$ on the right-hand side of (5) is the initial occupation probability of level $|i\rangle$, the initial off-diagonal elements of the density matrix being taken to be zero. The set of equations (5) constitute a set of N linear equations for the $\tilde{P}_i$ and the solution may be written down following the methods set out in Ref. 18. One obtains

$$\tilde{P}_i(z)=P_i(0)\mathscr{G}_i+\sum_j{}^{*} P_j(0)\mathscr{G}_j W_{ji}\mathscr{G}_i^{(j)} + \sum_{j,k}{}^{*} P_k(0)\mathscr{G}_k W_{kj}\mathscr{G}_j^{(k)} W_{ji}\mathscr{G}_i^{(k,j)}+\cdots\ , \tag{7}$$

the general term of the series being obvious by inspection. The asterisk on the sums indicates that all the subscripts in a given term must all be different from each other. Thus, in Eq. (7), $\sum_{j,k}^{*}$ stands for $\sum_{k\neq i,j}\sum_{j\neq i}$, etc. The $\mathscr{G}_i$'s, which may be considered to be propagators of a kind, are defined by the relations

$$\mathscr{G}_l^{(k,j,\ldots)}=(\mathscr{D}_l^{(k,j,\ldots)})^{-1}, \tag{8}$$

$$\mathscr{D}_l^{(k,j,\ldots)}=z+W_l-\sum_m{}^{*} W_{lm}\mathscr{G}_m^{(l,k,j\ldots)}W_{ml} + \sum_{m,n}{}^{*} W_{lm}\mathscr{G}_m^{(l,k,\ldots)}W_{mn}\mathscr{G}_n^{(m,l,k,\ldots)}W_{nl} + \cdots\ , \tag{9}$$

where the terms alternate in sign, and the asterisk on the sums has the same meaning as before except that, in addition, the variables summed over are not allowed to take on the values of the superscripts appearing in the summand. Thus the $\mathscr{G}$'s, being defined iteratively by Eqs. (8) and (9) have the form of a sum of products of continued fractions.

If we invert Eq. (7) we obtain

$$P_i(t)=P_i(0)U_i(t)+\sum_j{}^{*}\int_0^t dt_2\int_0^{t_2}dt_1 P_j(0)$$

$$\times U_j(t_1)W_{ji}(t_2-t_1)U_i(t-t_2)+\cdots\ , \quad (10)$$

where $U_i(t)$ is the inverse Laplace transform of $\mathcal{G}_i(z)$. This equation has a simple physical interpretation. The first term is the product of the probability, $P_i(0)$, of the system being in the state $|i\rangle$ at time $t=0$ with the probability $U_i(t)$ that the system remains in the state $|i\rangle$ for the duration t. The second term represents the probability that the system is in state $|j\rangle$ at time $t=0$ $[P_j(0)]$, times the probability that it remains in that state for an interval t_1 $[U_j(t_1)]$, times the probability that in the interval (t_2-t_1) it makes a transition from state $|j\rangle$ to state $|i\rangle$ $[W_{ji}(t_2-t_1)]$, times the probability that it propagates in state i for the time interval $(t-t_2)$ $[U_i(t-t_2)]$, integrated over all values of $t_1\le t_2$ and all $t_2\le t$, and summed over all intermediate states $|j\rangle$. Higher-order terms have a similar interpretation. Expression (7) is simpler to work with than expression (10) because, instead of convolutions, straightforward products appear in the former. We emphasize that the solutions (7)–(10) are exact.

We now consider the four-level model defined in Sec. II A. In this case the restrictions on the sums cause the sums to terminate after only a few terms. Assuming the initial conditions

$$P_1(0)=1,\ \ P_0(0)=P_2(0)=P_3(0)=0 \quad (11)$$

one obtains from Eq. (7) the solution (for $i=3$)

$$\tilde{P}_3\equiv\tilde{P}_3^{(1)}+\tilde{P}_3^{(0)}\ , \quad (12)$$

where

$$\tilde{P}_3^{(1)}\equiv\mathcal{G}_1(W_{13}+W_{12}\mathcal{G}_2^{(1)}W_{23})\mathcal{G}_3^{(1,2)}\ , \quad (13)$$

$$\tilde{P}_3^{(0)}\equiv\mathcal{G}_1W_{10}\mathcal{G}_0^{(1)}(W_{03}+W_{02}\mathcal{G}_2^{(0,1)}W_{23})\mathcal{G}_3^{(0,1,2)}$$
$$+\mathcal{G}_1W_{12}\mathcal{G}_2^{(1)}W_{20}\mathcal{G}_0^{(1,2)}W_{03}\mathcal{G}_3^{(0,1,2)}\ . \quad (14)$$

The terms in Eq. (13) represent the probability of a transition from state $|1\rangle$ to state $|3\rangle$ without the direct intervention of state $|0\rangle$; the terms in Eq. (14) on the other hand, represent the probability of a transition from state $|1\rangle$ to state $|3\rangle$, which proceeds through state $|0\rangle$ as an intermediate state. These processes are shown schematically in Fig. 2.

III. PROBE-FIELD APPROXIMATION

Some simplification can be made if we impose the *probe-field approximation*

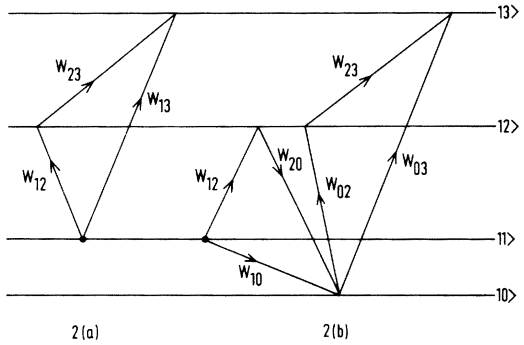

FIG. 2. Basic processes connection $|1\rangle$ with $|3\rangle$, divided into those which do (b) and do not (a) pass through state $|0\rangle$.

$$|V_{23}|\ll\Gamma_{12},\Gamma_{02}\ , \quad (15)$$

and assume that laser A is of *saturating intensity*

$$|V_{02}|\sim|V_{12}|\gg|V_{2,3}|,\Gamma_{i,j}\ \ (\text{all } i,j) \quad (16)$$

where

$$\Gamma_{ij}\equiv\tfrac{1}{2}(\gamma_i+\gamma_j)+\Delta_{ij} \quad (17)$$

and $\Delta_{01}=0$, $\Delta_{02}=\Delta_{12}=\Delta_a$, $\Delta_{03}=\Delta_{13}=\Delta_a+\Delta_b$, $\Delta_{23}=\Delta_b$, $\Delta_{ij}=\Delta_{ji}$. These are realistic assumptions for most of the experiments which have been carried out. Under these approximations we obtain reasonably simple expressions for the $W_{ij}(z)$ which are given in the Appendix.

For example, the one-photon contribution to $W_{12}(z)$, $W_{12}^{(1)}(z)$ is given by [Eq. (A5)]

$$W_{12}^{(1)}(z)=2\,\mathrm{Re}\left\{\frac{|V_{12}|^2}{z+\Gamma_{12}+i\delta_1+|V_{02}|^2/(z-iE_{1,0})}\right\}. \quad (18)$$

We have introduced the principal detunings $\delta_1\equiv\delta_{2,1}$, $\delta_2\equiv\delta_{3,2}$, where

$$\delta_1\equiv E_{2,1}-\omega_a,\ \ \delta_2\equiv E_{3,2}-\omega_b,\ \ E_{x,y}\equiv E_x-E_y\ . \quad (19)$$

All the other detunings δ_{ij} can be expressed in terms of δ_1, δ_2, and $E_{1,0}$. For example, $\delta_{30}=E_{1,0}+\delta_1+\delta_2$. In obtaining Eq. (18) we have made use of the inequalities

$$\omega_a\gg E_{1,0}\gg|V_{12}|\ , \quad (20)$$

which follow from the specification of our model in the introduction. By writing the denominator of Eq. (18) in the form

$$z+\Gamma_{12}+i\left[\delta_1+\frac{|V_{02}|^2E_{1,0}}{z^2+E_{1,0}^2}\right]+\frac{z|V_{02}|^2}{z^2+E_{1,0}^2}, \tag{21}$$

we see that it contains the complex light shift

$$S(z)=-i|V_{02}|^2/(z-iE_{1,0}). \tag{22}$$

Expression (22) is z dependent but in the steady state ($z\to 0$) it is purely real:

$$S_1(0)=|V_{02}|^2/E_{1,0}. \tag{23}$$

Setting $z=0$ in Eq. (21), which becomes

$$\Gamma_{12}+i[\delta_1+S_1(0)], \tag{24}$$

we see that $S_1(0)$ modifies the effective detuning in the steady state to $\delta_1+S_1(0)$.

Note that if we had calculated $W_{12}^{(1)}(z)$ to higher order we would have obtained the light shift

$$S_1(z)=-i|V_{02}|^2\{z-iE_{1,0}+|V_{12}|^2/[z+\Gamma_{02}-i(\delta_1+E_{1,0})]\}^{-1}. \tag{25}$$

This shows that the energy levels appearing in the denominator of the light shift are themselves light shifted. This can significantly affect the value of $S_1(0)$ if $|V_{12}|>>|E_{1,0}|$ (assuming $|\delta_1|,\Gamma_{02}<<|E_{1,0}|$). However, this is contrary to the conditions (20) we have assumed here, and we take $W_{12}^{(1)}$ to be given by Eq. (18).

IV. NUMERICAL RESULTS

Consider the situation where the saturating laser is tuned to near resonance with the $|1\rangle\leftrightarrow|2\rangle$ transition, and where all the population at $t=0$ is the $|1\rangle$ level. Population will be transferred from $|1\rangle$ to $|2\rangle$ (and back) by resonant stimulated processes, and thence from $|2\rangle$ to $|0\rangle$ by spontaneous and stimulated off-resonant processes. There is only a small probability of repopulating level $|2\rangle$ from level $|0\rangle$, as the laser is far from resonance with this transition, and we have assumed that the Rabi frequency is much smaller than the energy-level separation $E_{1,0}$. Thus, there is a largely irreversible transfer of population from $|1\rangle$ to $|0\rangle$ and a resulting change in the Autler-Townes spectrum with time. For short times the dominant contribution is the doublet arising essentially from the $|1\rangle\leftrightarrow|2\rangle\leftrightarrow|3\rangle$ subsystems, while for larger times the dominant contribution is the two-photon peak at $\delta_2=-E_{1,0}$ arising from the $|0\rangle\leftrightarrow|2\rangle\leftrightarrow|3\rangle$ subsystem. These features are evident in Fig. 3, where we take $E_{1,0}=1.0\times10^{10}$, $\delta_1=0$, $|V_{12}|=|V_{02}|=2\times10^9$, $V_{23}=2\times10^5$, $\gamma_{21}=\gamma_{20}=3\times10^6$, $\gamma_{32}=7\times10^5$, $\Delta_a=0$, $\Delta_b=2\times10^8$, all quantities being measured in Hz. The unit of time is $\gamma_2^{-1}=1.7\times10^{-7}$ s, and the vertical scale is logarithmic. These parameters have been chosen to enable the relative time development of the two-photon and doublet peaks to be followed conveniently on the same graph. At short times the doublet is the largest contribution, whereas at $\gamma_2 t=100$ (approximately the steady state) the two-photon peak is dominant. There is evidently a slight but definite change in the asymmetry of the doublet with time.

Now we choose a set of parameters which correspond roughly to the transitions (3^2S $F=1$ and $F=2\leftrightarrow(3^2P_{1/2}$ $F=2)\leftrightarrow(5^2S_{1/2}$ $F=2)$ of sodium. With all quantities being measured in MHz, we take $\gamma_{21}=\gamma_{20}=10^8$, $\gamma_{32}=6.6\times10^6$, and $E_{1,0}=1.1\times10^{10}$. We also choose modest laser intensities, $|V_{21}|=|V_{20}|=3.1\times10^8$, $|V_{32}|=6.3\times10^6$. In Fig. 4 we show the doublet part of the spectra for exact resonance ($\delta_1=0$) with zero laser linewidths, at the three instants, $T=(5,10,\text{and }100)\times3.9\times10^{-9}$ s. (The unit of time is now $|V_{12}|^{-1}$.) The two-photon peak is well separated from the doublet for these values of the parameters and has not been shown in the remaining figures. The most obvious feature of these spectra is that their asymmetry is markedly time dependent. At $|V_{21}|T=5$, the low-frequency peak is much larger than the high-frequency peak, whereas at $|V_{21}|T=100$, the spectrum is practically symmetric. (For these values of the parameters the steady-state light shift is small, 8.7×10^6, and we have effectively resonant excitation.) At the smaller times modulations of the spectra are apparent. Thus at $|V_{21}T|=5$ the high-frequency peak is split, and at $|V_{21}|T=5$, 10 there is a component at $\delta_2=0$, which vanishes for larger interaction times. We find that the steady state is reached for $|V_{21}|T\gtrsim200$, although for all $|V_{21}|T\gtrsim100$, the spectra are qualitatively similar, only the overall amplitude changing.

In Fig. 5, the parameters are the same as in Fig. 4 except that now $\Delta_a=3.1\times10^7$ Hz, $\Delta_b=6.3\times10^6$ Hz. The spectra are quite similar to those of Fig. 4, but the overall dependence of the asymmetry on the time is reduced. In both Figs. 5 and 4 there is

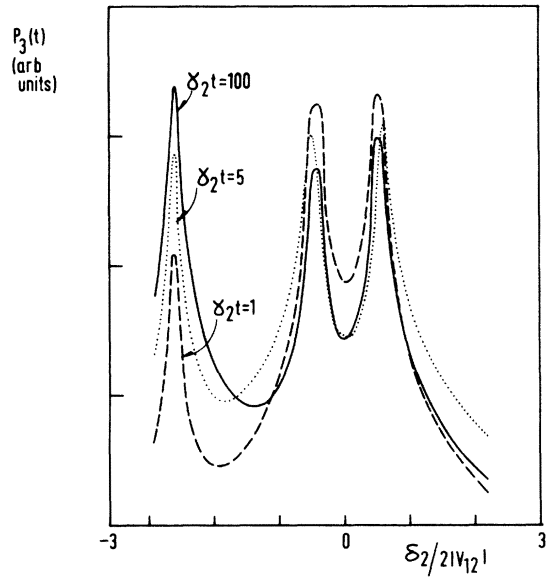

FIG. 3. Optical Autler-Townes spectrum [$P_3(t)$ against $\delta_2/2\,|\,V_{12}\,|\,$] at resonance ($\delta_1=0$) for the case $\gamma_{21}=\gamma_{20}=3\times10^6$, $\gamma_{32}=7\times10^5$, $\Delta_a=0$, $\Delta_b=2\times10^8$, $E_{1,0}=10^{10}$, $|\,V_{12}\,|=|\,V_{02}\,|=2\times10^9$, $|\,V_{23}\,|=2\times10^5$. (All quantities measured in Hz.)

a slight variation of the position of the high-frequency peak with time.

In Fig. 6 we have increased the value of the saturating laser linewidth Δ_a to $\Delta_a=10^8$ MHz, all other parameters being the same as in Fig. 4. This

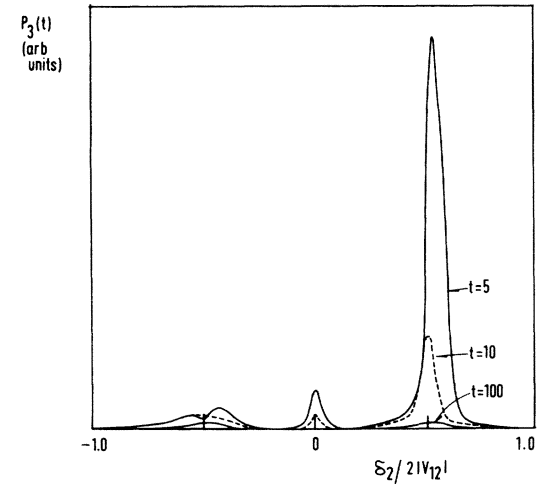

FIG. 4. Optical Autler-Townes spectrum [$P_3(t)$ plotted against $\delta_2/2\,|\,V_{12}\,|\,$] at resonance ($\delta_1=0$) for the case $\gamma_{21}=\gamma_{20}=10^8$, $\gamma_{32}=6.3\times10^6$, $|\,V_{20}\,|=|\,V_{21}\,|=3.1\times10^8$, $E_{1,0}=1.1\times10^{10}$, $|\,V_{32}\,|=6.3\times10^6$, $\Delta_a=\Delta_b=0$. (All quantities measured in Hz.)

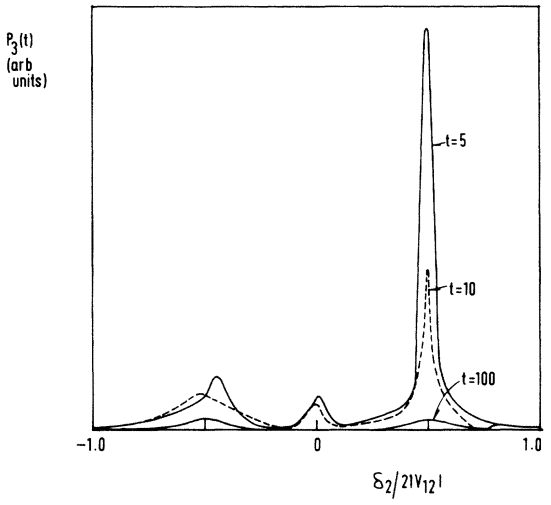

FIG. 5. As in Fig. 4 but with $\Delta_a=3.1\times10^7$, $\Delta_b=6.3\times10^6$ Hz.

single change produces a great change in the spectra. First of all the spectra for all three times are almost symmetric, and the asummetry does not change significantly with time. Secondly, the higher-frequency modulations are much more clearly seen at smaller times than in the previous two figures. Such high-frequency modulations were seen in the three-level problem discussed previously.[5]

The value of the saturating laser linewidth, $\Delta_a=\frac{1}{2}\gamma_2$, corresponds to the value at which the

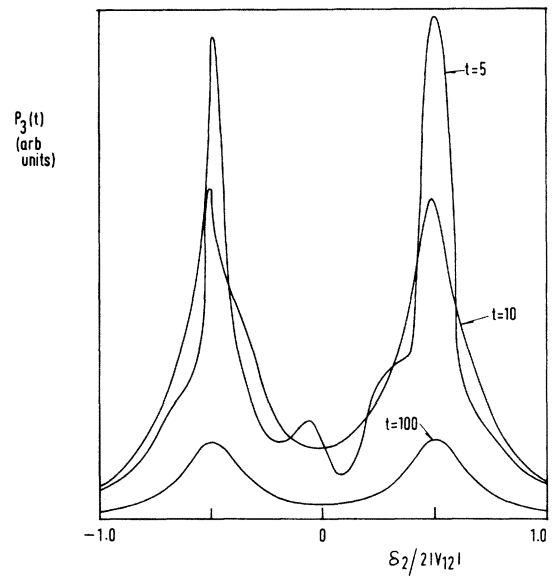

FIG. 6. As in Fig. 4 but with $\Delta_a=10^8$ Hz.

sign of the asymmetry changes in the steady state for the three-level systems, as pointed out by Georges and Lambropoulos.[13] Thus it is not surprising that the spectra are practically symmetry in this case.

In Fig. 7, the data is the same as in Fig. 4 except that a modest detuning, $\delta_1 = 10^8$ Hz, has been introduced. This is not sufficient to significantly alter the nature of the asymmetry in the steady state, but it radically alters the nature of the transient asymmetry. Thus for $|V_{21}|T=5,10$, the high-frequency peak is the larger, but for $|V_{21}|T=100$, the low-frequency peak is (marginally) the larger.

We have demonstrated that the asymmetry of the Autler-Townes spectra can vary markedly with time, and that it is strongly influenced by the laser linewidths. The nature of the asymmetry of the transient spectra can be qualitatively different from those of the steady state.

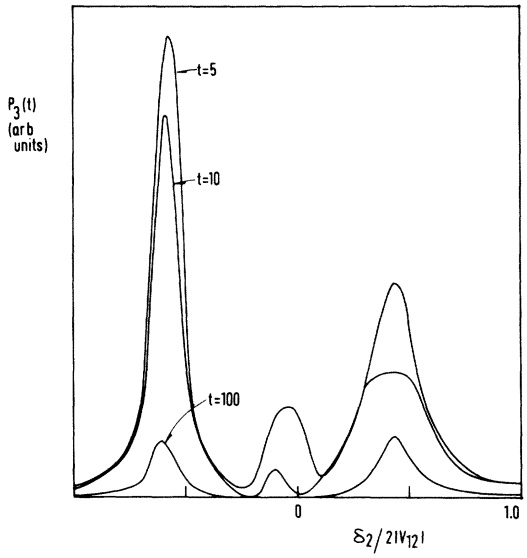

FIG. 7. As in Fig. 4 but with $\Delta_a = \Delta_b = 0$, $\delta_1 = 10^8$ Hz.

APPENDIX

For completeness, we present here explicit formulas for the rates, $W_{ij}(z)$. First of all we note the relations

$$W_{02} = W_{02}^{(1)} - W_{01} - W_{03} , \tag{A1}$$

$$W_{12} = W_{12}^{(1)} - W_{01} - W_{13} , \tag{A2}$$

$$W_{23} = W_{23}^{(1)} - W_{03} - W_{13} , \tag{A3}$$

where the $W_{ij}^{(1)}$ represent essentially one-photon contributions to the W_{ij}. Making use of the approximations (15), (16), and (20) we find

$$W_{02}^{(1)} = 2|V_{02}|^2 \mathrm{Re}[z+\Gamma_{02}+i(\delta_1+E_{1,0})+|V_{12}|^2/(z+iE_{1,0})]^{-1} , \tag{A4}$$

$$W_{12}^{(1)} = 2|V_{12}|^2 \mathrm{Re}[z+\Gamma_{12}+i\delta_1+|V_{02}|^2/(z-iE_{1,0})]^{-1} , \tag{A5}$$

$$\begin{aligned} W_{23}^{(1)} = 2|V_{23}|^2 \mathrm{Re}\{ & z+\Gamma_{23}+i\delta_2+|V_{12}|^2/[z+\Gamma_{13}-i(\delta_1+\delta_s)] \\ & +|V_{02}|^2/[z+\Gamma_{03}-i(\delta_1+\delta_2+E_{1,0})]\}^{-2} , \end{aligned} \tag{A6}$$

$$\begin{aligned} W_{01} = 2|V_{02}V_{21}|^2 \mathrm{Re}\{ & [z+\Gamma_{20}+i(\delta_1+E_{1,0})][(z+iE_{1,0})(z-i\delta_1+\Gamma_{12}) \\ & +|V_{12}|^2]+|V_{12}|^2(z+\Gamma_{12}-i\delta_1)\}^{-1} , \end{aligned} \tag{A7}$$

$$\begin{aligned} W_{03} = 2|V_{02}V_{23}|^2 \mathrm{Re}(& [z+\Gamma_{02}-i(\delta_1+E_{1,0})+|V_{12}|^2/(z-iE_{1,0})] \\ & \times\{(z+\Gamma_{23}-i\delta_2)[z+\Gamma_{03}-i(\delta_1+\delta_2+E_{1,0})]+|V_{02}|^2 \\ & +|V_{12}|^2[z+\Gamma_{03}-i(\delta_1+\delta_2+E_{1,0})]/[z+\Gamma_{13}-i(\delta_1+\delta_2)]\})^{-1} , \end{aligned} \tag{A8}$$

and finally

$$W_{13}=2\,|\,V_{12}V_{23}\,|^{2}\,\mathrm{Re}\{[z+\Gamma_{12}-i\delta_{1}+|\,V_{02}\,|^{2}/(z+iE_{1,0})]$$
$$\times\{(z+\Gamma_{23}-i\delta_{2})[z+\Gamma_{13}-i(\delta_{1}+\delta_{2})]+|\,V_{12}\,|^{2}$$
$$+\;|\,V_{02}\,|^{2}[z+\Gamma_{13}-i(\delta_{1}+\delta_{2})]/[z+\Gamma_{03}-i(\delta_{1}+\delta_{2}+E_{1,0})]\}\}^{-1}\,. \tag{A9}$$

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