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Time-dependent spectra of a strongly driven three-level atom

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Time-dependent features of the fluorescence radiation from a strongly driven three-level atom in a Raman configuration are studied applying the Eberly-Wódkiewicz counting-rate definition of the spectrum. Using our earlier formalism, where we assume that one or both of the driving fields are intense, we obtain analytical expressions for the time-dependent fluorescence spectra. We have also studied the influence on the transient spectrum of the initial preparation of the atom in three different dressed states.

I. INTRODUCTION

Recently there has been some interest in studying atom-field interaction dynamics in the framework of time-dependent (physical) spectra[1] (TDS). TDS takes into account the presence of the measuring device (e.g., a Fabry-Perot interferometer). Eberly *et al.*[2] have studied the temporal behavior of the fluorescence spectrum of a two-level atom irradiated by a strong monochromatic laser field which is switched on suddenly at an initial time. They have observed that the spectrum changes in time and the total spectral intensity oscillates according to the Rabi flopping frequency. The calculated transient spectrum retains its symmetric, three-peaked structure for a resonant driving field. For a strong off-resonant driving field, the temporal spectrum can be quite asymmetric before the steady state is reached. Even when the laser field is tuned to exact resonance, Lu *et al.*[3] have demonstrated that the symmetry of the spectrum about the driving field is broken if the atom is initially prepared in pure dressed states of the atom-field Hamiltonian. Also, irradiation by a noisy laser can produce spectra that, even in steady state, are closely similar to the earlier time transient spectra obtained from irradiation by a monochromatic laser.[2] Time-dependent resonance fluorescence from a two-level atom has also been studied theoretically by several others[4–11] and in recent experiments.[12] In particular, the spectrum of resonance fluorescence following the removal of a strong laser has been studied in detail by Huang *et al.*[11] A different aspect of time-dependent resonance fluorescence spectra can arise when an atom is irradiated by a strong pulsed laser light of finite pulse length in which the process of turning on and off the pulse is to be reflected in the corresponding spectrum.[6,7,13–16]

The steady-state spectrum of the fluorescent radiation from a three-level atom in the presence of one or more strong laser fields is known to show even more interesting features. These features depend on the detuning from the atomic-transition frequencies and the strength of the two laser fields driving the atom. For example, the steady-state fluorescence spectra from a three-level atom both in the cascade and Raman configurations contain five components which are symmetric about the spectral center at the corresponding laser frequencies when the external laser fields are resonant and monochromatic.[17–21] The corresponding spectra from an atom in the V configuration are, however, symmetric triplets.[22] Further, if the driving fields are detuned from the atomic transition frequencies, the spectra exhibit as many as seven Lorentzian peaks.[17,19] On the other hand, for example, in the cascade configuration, if the lower transition is driven by a strong field whereas the upper transition is probed by a weak field, the spectrum from the lower transition is found to be the Stark triplet which is characteristic of a strongly driven two-level atom.[23,24] The upper spectrum, in this case, is the so-called Autler-Townes doublet.[20,25,26] It has also been shown that the fluctuations in the laser fields affect the spectra considerably. For small phase fluctuations the central peaks and the sidebands are broadened and decreased in intensity as the laser bandwidths are increased. For large fluctuations, the Stark multiplets tend to reduce to Stark doublets.[27–30]

Transient spectra for a three-level atom exposed to a strong driving field and a weak probe field have been recently studied both theoretically[31] and experimentally.[32] As expected, the transient probe spectrum ultimately evolves to the well-known steady-state Autler-Townes doublet. However, the transient spectra are dramatically different from those found in the steady state, depending on whether the atom was initially prepared in a pure dressed state or not. Even the preparation of the pure dressed states using amplitude and phase-controlled resonant excitation fields has been experimentally demonstrated.[33]

In this paper we discuss the transient spectra of a three-level atom in a Raman configuration interacting with two strong near-resonant laser fields that are switched on at $t=0$. In Sec. II we formulate the problem. In Sec. III we obtain analytical solution for the TDS in the high-field approximation. The present formulation can be extended to study the effect of fluctuations in the driving field on TDS. This is being studied and will be presented elsewhere.

II. FORMULATION OF THE PROBLEM

A. Master equation

We consider a three-level atom with unequally spaced levels $(E_1 > E_2 > E_3)$ in the Raman configuration, interacting with two single-mode laser fields (Fig. 1). The first laser driving the Rayleigh transition from the ground state $|3\rangle$ to the excited state $|1\rangle$ has a frequency Ω_1 which is detuned from the atomic transition frequency $\omega_1 = E_1 - E_3$ by an amount Δ_1. The second laser of frequency Ω_2 drives the Stokes transition $|2\rangle$ to $|1\rangle$ and is detuned from the atomic transition frequency $\omega_2 = E_1 - E_2$ by an amount Δ_2. The dynamics of such a system is described by the master equation[34] for the reduced density operator ρ

$$\frac{d\rho(t)}{dt} = -i[H_0,\rho(t)] - \gamma_1[A_{11}\rho(t) + \rho(t)A_{11} - 2A_{31}\rho(t)A_{13}] - \gamma_2[A_{11}\rho(t) + \rho(t)A_{11} - 2A_{21}\rho(t)A_{12}]$$
$$- \gamma_3[A_{22}\rho(t) + \rho(t)A_{22} - 2A_{32}\rho(t)A_{23}] - \gamma_4[A_{33}\rho(t) + \rho(t)A_{33} - 2A_{23}\rho(t)A_{32}] \ , \qquad (2.1)$$

where

$$H_0 = \alpha_1(A_{13} + A_{31}) + \alpha_2(A_{12} + A_{21}) + \Delta_1 A_{11} + \Delta A_{22} \ , \qquad (2.2a)$$

$$\Delta = \Delta_1 - \Delta_2, \quad \alpha_j = d_j E_j(t)/2 \ . \qquad (2.2b)$$

The master equation (2.1) involves the usual electric dipole and rotating-wave approximations. Further, the Born and Markov approximations with respect to the interaction with the continuum modes of the radiation field are inherent. Lastly, the equation is written in a frame rotating with respect to the laser frequencies. In (2.1) the coefficients $2\gamma_1$ and $2\gamma_2$ are the radiative spontaneous transition probabilities per unit time for the atom to make a Rayleigh and Stokes transition, respectively. $2\gamma_3$ and $2\gamma_4$ are the nonradiative relaxations between the dipole-forbidden transitions $|2\rangle$ to $|3\rangle$ and $|3\rangle$ to $|2\rangle$, respectively, which are added phenomenologically to obtain the nonvanishing steady-state resonance spectrum.[21] $2\alpha_1$ and $2\alpha_2$ in (2.2) represent the Rabi frequencies corresponding to the Rayleigh and Stokes transition, respectively, d_j $(j=1,2)$ are the dipole matrix elements, and $E_j(t)$ are the driving field amplitudes which we consider to be constant. The operators $A_{mn} = |m\rangle\langle n|$ obey the usual commutation relations

$$[A_{mn}, A_{pq}] = A_{mq}\delta_{np} - a_{pn}\delta_{qm} \qquad (2.3a)$$

and the closure property

$$A_{11} + A_{22} + A_{33} = 1 \ . \qquad (2.3b)$$

We might mention here that by taking the matrix elements of ρ between the atomic states $|i\rangle$ and $|j\rangle$ $(i,j=1,2,3)$ one obtains from the master equation (2.1), the familiar Bloch equations. The Bloch equations with the various relaxation mechanisms have been extensively used in the three-level spectroscopic studies.[35,36] Also, master equations for multilevel systems have been introduced in connection with the theory of masers by Fain and Khanin.[37]

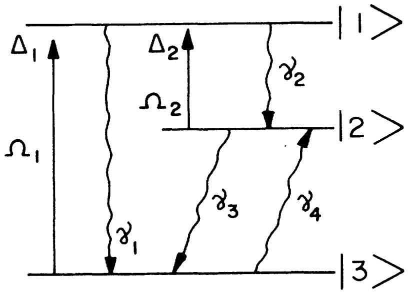

FIG. 1. Schematic diagram of a three-level atom in a Raman configuration interacting with two monochromatic fields.

B. Dressed atomic operators and the secular approximation.

The master equation (2.1) may be directly used to compute the one-time expectation values of the atomic operators numerically.[27] However, to obtain analytical expressions we assume that the fields are strong. In this limit we can also obtain a steady-state solution for the atomic density operator. We follow essentially the method of Ref. 29.

We first diagonalize the Hamiltonian H_0 of the driven atomic system given in Eq. (2.2). Denoting the $|\psi_i\rangle$ the eigenstates of H_0 corresponding to the eigenvalue $-p_j$, we express the atomic states $|i\rangle$ by

$$|1\rangle = \sum_{i=1}^{3} a_i |\psi_i\rangle \ , \qquad (2.4a)$$

$$|2\rangle = \sum_{i=1}^{3} b_i |\psi_i\rangle \ , \qquad (2.4b)$$

$$|3\rangle = \sum_{i=1}^{3} c_i |\psi_i\rangle \ , \qquad (2.4c)$$

where a_i, b_i, and c_i have the explicit expressions

$$a_i=-p_i\alpha_2 N_i \ ,$$
$$b_i=(p_i^2+p_i\Delta_1-\alpha_1^2)N_i \ , \qquad (2.5)$$
$$c_i=\alpha_1\alpha_2 N_i \ ,$$

where the constants N_i are defined as

$$N_i=[p_i^2\alpha_2^2+(p_i^2+p_i\Delta_1-\alpha_1^2)^2+\alpha_1^2\alpha_2^2]^{-1/2} \qquad (2.6)$$

and p_i are the roots of the cubic equation

$$p^3+p^2(\Delta_1+\Delta)-p(\Omega^2-\Delta_1\Delta)-\alpha_1^2\Delta=0 \ , \qquad (2.7)$$

where

$$\Omega=(\alpha_1^2+\alpha_2^2)^{1/2} \ . \qquad (2.8)$$

The cubic equation can be solved readily whenever $\Delta_1=\Delta_2$. In this case the coefficients a_i, b_i, and c_i can be written down explicitly.[22]

We may now express the original operators A_{ij} appearing in the master equation (2.1) in terms of the new dressed operators $B_{ij}=|\psi_i\rangle\langle\psi_j|$ by means of the relations

$$A_{11}=\sum_{k,l=1}^{3} a_k a_l B_{kl} \ , \qquad (2.9a)$$

$$A_{12}=\sum_{k,l=1}^{3} a_k b_l B_{kl}=A_{21}^{\dagger} \ , \qquad (2.9b)$$

$$A_{13}=\sum_{k,l=1}^{3} a_k c_l B_{kl}=A_{31}^{\dagger} \ , \qquad (2.9c)$$

$$A_{22}=\sum_{k,l=1}^{3} b_k b_l B_{kl} \ , \qquad (2.9d)$$

$$A_{23}=\sum_{k,l=1}^{3} b_k c_l B_{kl}=A_{32}^{\dagger} \ , \qquad (2.9e)$$

$$A_{33}=\sum_{k,l=1}^{3} c_k c_l B_{kl} \ . \qquad (2.9f)$$

The reciprocal relations between B_{ij} and A_{ij} can be easily written down by noting that the transformation matrix involving the products of a_i, b_i, and c_i is real and orthogonal. The new operators B_{ij} obviously satisfy the same commutation relations as the old. Also, in terms of the new operators the Hamiltonian H_0 takes the simple form

$$H_0=-\sum_{k=1}^{3} p_k B_{kk} \ , \qquad (2.10)$$

and under the action of the Hamiltonian H_0, $B_{ij}(t)$ evolves with time as

$$B_{ij}(t)=B_{ij}(0)\exp[-i(p_i-p_j)t] \ . \qquad (2.11)$$

Next, we go over to the interaction representation by defining

$$\tilde{\rho}(t)=\exp\left[-it\sum_k p_k B_{kk}\right]\rho(t)\exp\left[it\sum_k p_k B_{kk}\right] \ , \qquad (2.12)$$

whereby the resulting master equation for $\tilde{\rho}(t)$ splits into two parts, viz, the one containing no oscillatory terms and the other containing rapidly oscillatory terms, such as $\exp[\pm i(p_i-p_j)t]$, $\exp[\pm 2i(p_i-p_j)t]$, and $\exp[\pm i(2p_i-p_j-p_k)t]$ $(i,j,k=1,2,3;\ i\neq j)$. Making the secular approximation, that is, neglecting the oscillatory terms and finally reverting back to the Schrödinger picture, we arrive at the master equation

$$\frac{d\rho(t)}{dt}=i\sum_k p_k[B_{kk},\rho(t)]-\sum_k g_k'[B_{kk}\rho(t)+\rho(t)B_{kk}]$$
$$+2\sum_{k,l} e_{kl}B_{kk}\rho(t)B_{ll}+2\sum_{\substack{k,l\\(k\neq l)}} f_{kl}B_{kl}\rho(t)B_{lk} \ , \qquad (2.13)$$

where g_k', e_{kl}, and f_{kl} are given by

$$g_k'=(\gamma_1+\gamma_2)a_k^2+\gamma_3 b_k^2+\gamma_4 c_k^2 \ , \qquad (2.14)$$

$$e_{kl}=(\gamma_1 c_k c_l \gamma_2 b_k b_l)a_k a_l+(\gamma_3+\gamma_4)b_k b_l c_k c_l$$
$$=e_{lk} \ , \qquad (2.15)$$

$$f_{kl}=(\gamma_1 c_k^2+\gamma_2 b_k^2)a_l^2+\gamma_3 c_k^2 b_l^2+\gamma_4 b_k^2 c_l^2$$
$$(k\neq l) \ . \qquad (2.16)$$

Equation (2.13) has a steady-state solution[28,29]

$$\rho_{\mathrm{SS}}=D^{-1}\exp[-(\mu_1 B_{11}+\mu_2 B_{22})] \ , \qquad (2.17)$$

where the subscript SS denotes the steady state and μ_1 and μ_2 have the following expressions:

$$\mu_1=\ln[(f_{11}f_{22}-f_{12}f_{21})/(f_{13}f_{22}+f_{12}f_{23})] \ , \qquad (2.18)$$

$$\mu_2=\ln[(f_{11}f_{22}-f_{12}f_{21})/(f_{11}f_{23}+f_{13}f_{21})] \ . \qquad (2.19)$$

The quantities f_{ij} $(i\neq j)$ are defined in (2.16), while the constants f_{kk} $(k=1,2,3)$ are given by

$$f_{kk}=g_k'-e_{kk}=[\gamma_1(1-c_k^2)+\gamma_2(1-b_k^2)]a_k^2$$
$$+\gamma_3(1-c_k^2)b_k^2+\gamma_4(1-b_k^2)c_k^2 \ . \qquad (2.20)$$

The normalization factor D is chosen such that $\mathrm{Tr}\rho_{\mathrm{SS}}=1$ and can be easily shown to be

$$D=\mathrm{Tr}\{\exp[-(\mu_1 B_{11}+\mu_2 B_{22})]\}$$
$$=1+\exp(-\mu_1)+\exp(-\mu_2)$$
$$=[(f_{11}+f_{13})(f_{22}+f_{23})$$
$$-(f_{12}-f_{13})(f_{21}-f_{23})]/(f_{11}f_{22}-f_{12}f_{21}) \ . \qquad (2.21)$$

The solution (2.17) is useful for obtaining the steady-state expectation values of the dressed operators. It is clear from the nature of this solution that the expectation values are given by

$$\langle B_{ij}\rangle_{\mathrm{SS}}=\mathrm{Tr}(B_{ij}\rho_{\mathrm{SS}})=0 \quad (i\neq j) \ , \qquad (2.22)$$

$$\langle B_{ii}\rangle_{\mathrm{SS}}=\mathrm{Tr}(B_{ii}\rho_{\mathrm{SS}})$$
$$=(e^{-\mu_1}\delta_{i1}+e^{-\mu_2}\delta_{i2}+\delta_{i3})/D \ . \qquad (2.23)$$

The steady-state expectation values of the atomic operator A_{ij} follow from Eqs. (2.9), (2.22), and (2.23).

It may be added here that the case of a three-level atom in a strong external field has been previously treated by Fain and Khanin.[38] Their treatment includes the incoherent, steady-state pumping mechanism for the two excited states besides the external field. Steady-state values of the elements of the atomic density matrix were derived.[38] In the present paper, we have ignored the incoherent excitation mechanisms. The only excited mechanisms are the external fields and only damping present is natural radiative decay. The role of the nonradiative damping terms γ_3 and γ_4 is merely to prevent population trapping which occurs when the two external fields are in exact resonance with the respective transitions. Finally, we mention here that the approximate master equation (2.1) is valid under the conditions that the Rabi frequency $\alpha_1, \alpha_2 \gg \gamma_1, \gamma_2, \gamma_3, \gamma_4$.

III. TIME-DEPENDENT FLUORESCENT SPECTRA

A. One-time atomic operator averages

For an atomic operator O, we define the average as

$$\langle O \rangle = \mathrm{Tr}(O\rho) \ . \tag{3.1}$$

The general equation of motion for $\langle B_{ij}(t) \rangle$ can be readily obtained from (2.13) and reads as

$$\frac{d\langle B_{ij} \rangle}{dt} = -(i\Omega_{ij} + \Gamma_{ij})\langle B_{ij} \rangle + 2 \sum_{\substack{k,l \\ (k \neq l)}} f_{kl} \langle B_{ll} \rangle \delta_{ik} \delta_{jk} \ , \tag{3.2}$$

where

$$\Omega_{ij} = p_i - p_j \ , \tag{3.3}$$

$$\Gamma_{ij} = g_i' + g_j' - 2e_{ij} \ . \tag{3.4}$$

In particular, when $i = j$ we have the three coupled equations,

$$\frac{d\langle B_{11} \rangle}{dt} = -2f_{11}\langle B_{11} \rangle + 2f_{12}\langle B_{22} \rangle + 2f_{13}\langle B_{33} \rangle \ , \tag{3.5a}$$

$$\frac{d\langle B_{22} \rangle}{dt} = 2f_{21}\langle B_{11} \rangle - 2f_{22}\langle B_{22} \rangle + 2f_{23}\langle B_{33} \rangle \ , \tag{3.5b}$$

$$\frac{d\langle B_{33} \rangle}{dt} = 2f_{31}\langle B_{11} \rangle + 2f_{32}\langle B_{22} \rangle - 2f_{33}\langle B_{33} \rangle \ , \tag{3.5c}$$

and for $i \neq j$, we obtain the six equations,

$$\frac{d\langle B_{ij}(t) \rangle}{dt} = -(i\Omega_{ij} + \Gamma_{ij})\langle B_{ij}(t) \rangle \ . \tag{3.6}$$

Equations (3.5) and (3.6) can be solved and the solutions can be written in the compact form

$$\langle B_{ii}(t'+\tau) \rangle = \frac{g_i}{\nu_1 \nu_2} + \sum_{\nu=\nu_1,\nu_2} \frac{1}{\nu^2 - \nu_1 \nu_2} \left[(\nu^2 - 2h_i \nu + g_i)\langle B_{ii}(t') \rangle + \sum_{j(\neq i)} (-2f_{ij}\nu + g_i)\langle B_{jj}(t') \rangle \right] \exp(-\nu\tau) \ , \tag{3.7}$$

$$\langle B_{ij}(t'+\tau) \rangle = \langle B_{ij}(t') \rangle \exp[-(i\Omega_{ij} + \Gamma_{ij})\tau] \quad (i \neq j) \ , \tag{3.8}$$

where

$$h_1 = f_{13} + f_{22} + f_{23} \ , \tag{3.9a}$$

$$h_2 = f_{11} + f_{13} + f_{23} \ , \tag{3.9b}$$

$$h_3 = f_{11} + f_{22} \ , \tag{3.9c}$$

$$g_1 = 4(f_{13}f_{22} + f_{23}f_{12}) \ , \tag{3.10a}$$

$$g_2 = 4(f_{11}f_{23} + f_{13}f_{21}) \ , \tag{3.10b}$$

$$g_3 = 4(f_{11}f_{22} - f_{12}f_{21}) \ , \tag{3.10c}$$

and ν_1 and ν_2 are the two roots of the quadratic equation

$$\nu^2 - (h_1 + h_2 + h_3)\nu + (g_1 + g_2 + g_3) = 0 \ . \tag{3.11}$$

From Eq. (3.7) and (3.8) it is clear that the steady-state solutions for the expectation values of B_{ij} are as follows:

$$\langle B_{ij} \rangle_{\mathrm{SS}} = g_i / \nu_1 \nu_2 \quad (i \neq j) \ , \tag{3.12a}$$

$$\langle B_{ii} \rangle_{\mathrm{SS}} = 0 \ . \tag{3.12b}$$

B. Time-dependent fluorescence spectra

The time-dependent fluorescence spectra can be written as a double convolution integral of the form[1]

$$I_{R,S}(D,t,\Gamma) = 2\Gamma \,\mathrm{Re} \int_0^t dt' \exp[-\Gamma(t-t')] \int_0^{t-t'} d\tau \exp[(\Gamma/2 - iD)\tau] G_{R,S}(t',\tau) \ . \tag{3.13}$$

The subscript R (S) stands for the Rayleigh (Stokes) spectrum. In (3.13), Γ is the full width at half maximum of the (effectively Lorentzian) transmission peak of the Fabry-Perot interferometer used to measure the frequency spectrum of the fluorescence, D is the detuning or the frequency offset of the Fabry-Perot line center above the laser frequency, i.e., $D = \omega - \Omega_1$ ($\omega - \Omega_2$) for the Rayleigh (Stokes), and the autocorrelation functions $G_{R,S}(t',\tau)$ are given by

$$G_R(t',\tau)=\langle A_{13}(t'+\tau)A_{31}(t')\rangle \ , \tag{3.14a}$$

$$G_S(t',\tau)=\langle A_{12}(t'+\tau)A_{21}(t')\rangle \ . \tag{3.14b}$$

We note that

$$\langle A_{13}(t'+\tau)\rangle=\sum_i a_i c_i \langle B_{ii}(t'+\tau)\rangle + \sum_{\substack{i,j\\(i\neq j)}} a_i c_j \langle B_{ij}(t'+\tau)\rangle \ , \tag{3.15}$$

$$\langle A_{12}(t'+\tau)\rangle=\sum_i a_i b_i \langle B_{ii}(t'+\tau)\rangle + \sum_{\substack{i,j\\(i\neq j)}} a_i b_j \langle B_{ij}(t'+\tau)\rangle \ , \tag{3.16}$$

where the solutions for the expectation values $\langle B_{ij}(t'+\tau)\rangle$ can be written down from Eqs. (3.7) and (3.8). Expressing $A_{13}(t')$ and $A_{21}(t')$ also in terms of $B_{ij}(t')$ and applying the quantum regression theorem we obtain the following expression for $G_R(t',\tau)$ and $G_S(t',\tau)$:

$$G_{R,S}(t',\tau)=\sum_{i,j,l}\frac{a_i a_l \theta_i \theta_j g_i}{\nu_1\nu_2}\langle B_{jl}(t')\rangle + \sum_{\nu=\nu_1,\nu_2}\sum_{i,l}\frac{a_i a_l \theta_i}{\nu^2-\nu_1\nu_2}[\theta_i(\nu^2-2h_i\nu+g_i)\langle B_{il}(t')\rangle + \sum_{\substack{j\\(i\neq j)}}\theta_j(-2f_{ij}\nu+g_i)\langle B_{il}(t')\rangle]\exp(-\nu\tau) + \sum_{\substack{i,j,l\\(i\neq j)}} a_i a_l \theta_i \langle B_{il}(t')\rangle \exp[-(i\Omega_{ij}+\Gamma_{ij})\tau] \ , \tag{3.17}$$

$$\theta_i=\begin{cases} c_i & (\text{Rayleigh}) \ , \\ b_i & (\text{Stokes}) \ , \end{cases} \tag{3.18}$$

substituting once again the solutions for $\langle B_{ij}(t')\rangle$ from Eqs. (3.7) and (3.8) in Eq. (3.17) we can obtain an explicit expression for $G_{R,S}(t',\tau)$. Using this expression for $G_{R,S}(t',\tau)$ in (3.12) and noting that the τ and t' dependences of $G_{R,S}$ are both exponential, the integrals can be easily carried out. After some straightforward but tedious algebra we obtain the following explicit expressions for the spectra:

$$I_{R,S}(D,t,\Gamma)=\frac{A(D,t,\Gamma)}{(\Gamma/2)^2+D^2}+\sum_{n=1}^{4}S_n(D,t,\Gamma)+\sum_{n=1}^{3}U_n(D,t,\Gamma) \ , \tag{3.19}$$

where

$$A(D,t,\Gamma)=2\Gamma\sum_{\nu=0,\nu_1,\nu_2}B(\nu)f(\nu,D,t,\Gamma)\exp(-\nu t) \ , \tag{3.20}$$

$$B(\nu)=\sum_{k,l}a_k a_l c_k c_l \langle B_{kk}\rangle_{\text{SS}} Q_l(\nu) \ , \tag{3.21a}$$

$$f(\nu,D,t,\Gamma)=Z_1\{1-\exp[-(\Gamma/2-\nu)t]\cos(Dt)\}+Z_2\exp[-(\Gamma/2-\nu)t]\sin(Dt)-Z_3\{1-\exp[-(\Gamma-\nu)t]\} \ , \tag{3.21b}$$

$$Q_l(\nu)=\begin{cases}\langle B_{ll}\rangle_{\text{SS}}, & \nu=0\\ \langle B_{ll}(0)\rangle F_l(\nu)+\sum\limits_{\substack{j\\(j\neq l)}}\langle B_{jj}(0)\rangle G_{lj}(\nu), & \nu\neq 0\end{cases} \tag{3.21c}$$

$$Z_1=\frac{(\Gamma/2)(\Gamma/2-\nu)+D^2}{(\Gamma/2-\nu)^2+D^2} \ , \tag{3.21d}$$

$$Z_2=\frac{\nu D}{(\Gamma/2-\nu)^2+D^2} \ , \tag{3.21e}$$

$$Z_3=\frac{\Gamma}{2(\Gamma-\nu)} \ , \tag{3.21f}$$

$$F_l(\nu)=\frac{\nu^2-2h_l\nu+g_l}{\nu^2-\nu_1\nu_2} \ , \tag{3.21g}$$

$$G_{lj}(\nu)=\frac{-2f_{lj}\nu+g_l}{\nu^2-\nu_1\nu_2} \ , \tag{3.21h}$$

$$S_1(D,t,\Gamma)=\sum_{k}\sum_{\nu=\nu_1,\nu_2}\frac{a_k c_k W_{1k}(\nu)R_1(D,t,\Gamma,k,l,\nu_i,\nu_j)}{(\Gamma/2-\nu)^2+D^2}\ , \tag{3.22a}$$

$$S_2(D,t,\Gamma)=\sum_{k}\sum_{i,j=1}^{2}\frac{a_k c_k W_{2k}(\nu_i,\nu_j)R_2(D,t,\Gamma,k,l,\nu_i,\nu_j)}{(\Gamma/2-\nu_i)^2+D^2}\ , \tag{3.22b}$$

$$S_3(D,t,\Gamma)=\sum_{\substack{kl\\(k\neq l)}}\sum_{\nu=\nu_1,\nu_2}\frac{a_k^2 c_l^2 Q_k(\nu)R_3(D,t,\Gamma,k,l,\nu_i,\nu_j)}{(\Gamma/2-\Gamma_{kl})^2+(D+\Omega_{kl})^2}\ , \tag{3.22c}$$

$$S_4(D,t,\Gamma)=\sum_{\substack{k,l\\(k\neq l)}}\frac{a_k^2 c_l^2 \langle B_{kk}\rangle_{\mathrm{SS}}R_4(D,t,\Gamma,k,l,\nu_i,\nu_j)}{(\Gamma/2-\Gamma_{kl})^2+(D+\Omega_{kl})^2}\ , \tag{3.22d}$$

$$U_1(D,t,\Gamma)=\sum_{\substack{i,j\\(i\neq j)}}\sum_{\nu=\nu_1,\nu_2}\frac{a_j c_i \langle B_{ij}(0)\rangle W_{3i}(\nu)T_1(D,t,\Gamma,\nu,k,l,i,j)}{(\Gamma/2-\nu)^2+D^2}\ , \tag{3.23a}$$

$$U_2(D,t,\Gamma)=\sum_{i}\sum_{\substack{k,l\\(k\neq l)}}\frac{a_i c_i a_l c_k \langle B_{ii}\rangle_{\mathrm{SS}}\langle B_{kl}(0)\rangle T_2(D,t,\Gamma,\nu,k,l,i,j)}{(\Gamma/2)^2+D^2}\ , \tag{3.23b}$$

$$U_3(D,t,\Gamma)=\sum_{\substack{j,k,l\\(k\neq l,\ k\neq j)}}\frac{a_j a_k c_l^2 \langle B_{kj}(0)\rangle T_3(D,t,\Gamma,\nu,k,l,i,j)}{(\Gamma/2-\Gamma_{kl})^2+(D+\Omega_{kl})^2}\ , \tag{3.23c}$$

where

$$W_{1k}(\nu)=a_k c_k \langle B_{kk}\rangle_{\mathrm{SS}}F_k(\nu)+\sum_{\substack{l\\(l\neq k)}} a_l c_l \langle B_{ll}\rangle_{\mathrm{SS}}G_{kl}(\nu)\ , \tag{3.24a}$$

$$W_{2k}(\nu_i,\nu_j)=a_k c_k Q_k(\nu_j)F_k(\nu_i)+\sum_{\substack{l\\(l\neq k)}} a_l c_l Q_l(\nu_j)G_{kl}(\nu_i)\ , \tag{3.24b}$$

$$W_{3k}(\nu)=a_k c_k F_k(\nu)+\sum_{\substack{l\\(l\neq k)}} a_l c_l G_{kl}(\nu)\ , \tag{3.24c}$$

$$\begin{aligned}R_n(D,t,\Gamma,k,l,\nu_i,\nu_j)&\\ =2\Gamma(Y_{1n}\{1-\exp[-(\Gamma/2+\alpha_n-\beta_n)t]\cos(D+\eta_n)t\}&-Y_{2n}\{\exp[-(\Gamma/2+\alpha_n-\beta_n)t]\sin(D+\eta_n)t\}\\ &-Y_{3n}(1-\exp[-(\Gamma-\beta_n)t]))\exp(-\beta_n t)\quad(n=1,2,3,4)\ ,\end{aligned} \tag{3.25}$$

$$Y_{1n}=\frac{(\Gamma/2-\alpha_n)(\Gamma/2+\alpha_n-\beta_n)+(D+\eta_n)^2}{(\Gamma/2+\alpha_n-\beta_n)^2+(D+\eta_n)^2}\ , \tag{3.26a}$$

$$Y_{2n}=\frac{(2\alpha_n-\beta_n)(D+\eta_n)}{(\Gamma/2+\alpha_n-\beta_n)^2+(D+\eta_n)^2}\ , \tag{3.26b}$$

$$Y_{3n}=\frac{(\Gamma/2-\alpha_n)}{(\Gamma-\beta_n)}\ , \tag{3.26c}$$

with

$$\alpha_1=\beta_3=\nu,\quad \beta_1=\eta_1=\eta_2=\beta_4=0,\quad \alpha_2=\nu_i,\quad \beta_2=\nu_j,\quad \alpha_3=\alpha_4=\Gamma_{kl},\quad \eta_3=\eta_4=\Omega_{kl}\ , \tag{3.27}$$

$$\begin{aligned}T_n(D,t,\Gamma,\nu,k,l,i,j)=2\Gamma\{&(X_{1n}-X_{2n})\cos(\xi_n t)+(X_{3n}-X_{4n})\sin(\xi_n t)\\ &-\exp[-(\Gamma/2+\delta_n-\epsilon_n)t][X_{1n}\cos(D+\mu_n)t+X_{3n}\sin(D+\mu_n)t]\\ &+\exp[-(\Gamma-\epsilon_n)t]X_{2n}\}\exp(-\epsilon_n t)\ ,\end{aligned} \tag{3.28}$$

$$X_{1n}=\frac{(\Gamma/2+\Delta_n-\epsilon_n)(\Gamma/2-\delta_n)+(D+\mu_n)(D+\mu_n-\xi_n)}{(\Gamma/2+\delta_n-\epsilon_n)^2+(D+\mu_n-\xi_n)^2}\ , \tag{3.29a}$$

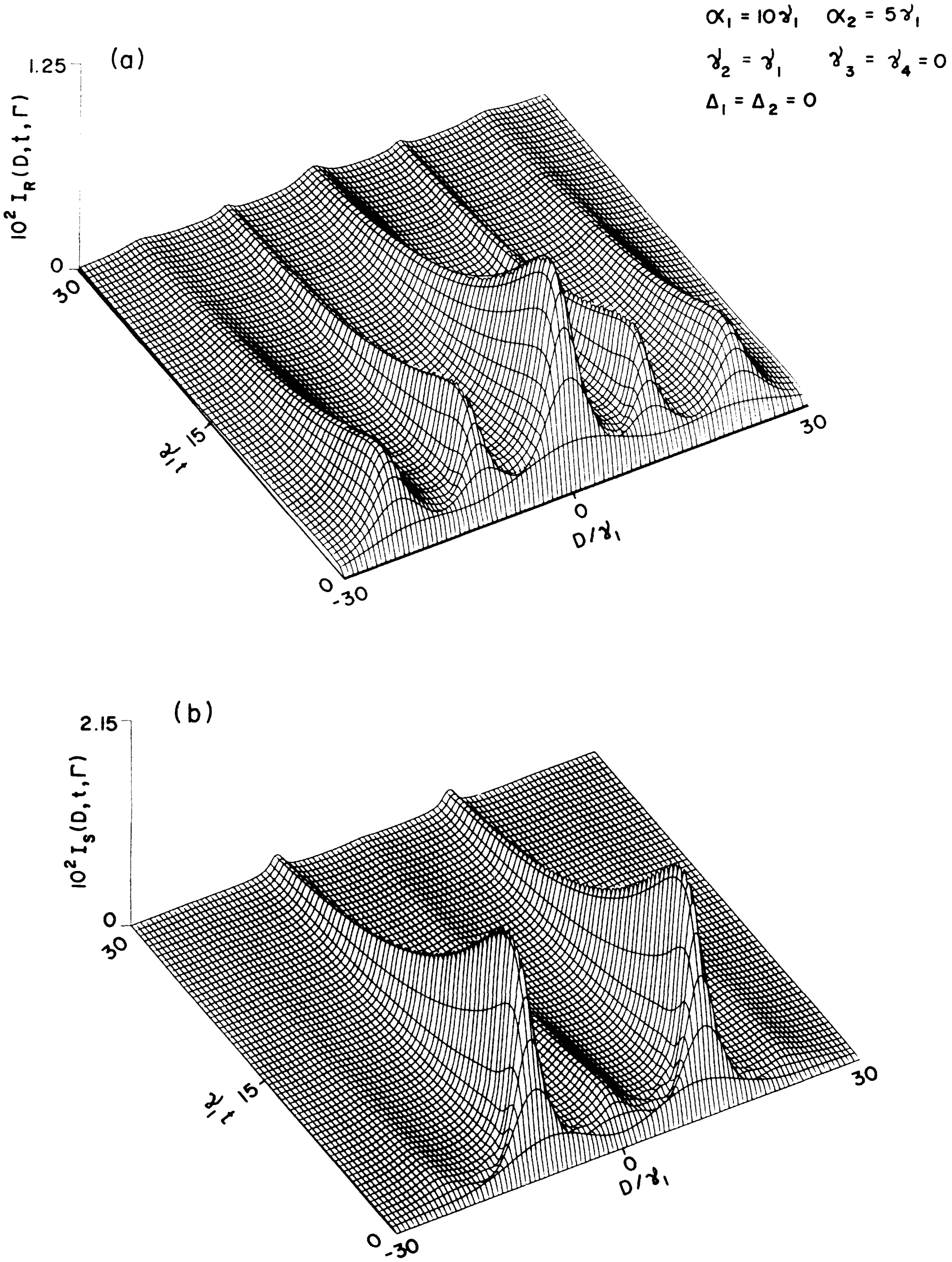

FIG. 2. Time-dependent (a) Rayleigh and (b) Stokes spectra as functions of time t and spectral frequency offset D under resonance conditions with $\gamma_3=\gamma_4=0$, the spectrometer bandwidth $\Gamma=0.1$, and the atom prepared initially in the ground state.

$$X_{2n}=\frac{(\Gamma/2-\delta_n)(\Gamma-\epsilon_n)-\xi_n(D+\mu_n)}{(\Gamma-\epsilon_n)^2+\xi_n^2}\,, \tag{3.29b}$$

$$X_{3n}=\frac{(\Gamma/2+\delta_n-\epsilon_n)(D+\mu_n)-(\Gamma/2-\delta_n)(D+\mu_n-\xi_n)}{(\Gamma/2+\delta_n-\epsilon_n)^2+(D+\mu_n-\xi_n)^2}\,, \tag{3.29c}$$

$$X_{4n}=\frac{(D+\mu)(\Gamma-\epsilon_n)+\xi_n(\Gamma/2-\delta_n)}{(\Gamma-\epsilon_n)^2+\xi_n^2}\,, \tag{3.29d}$$

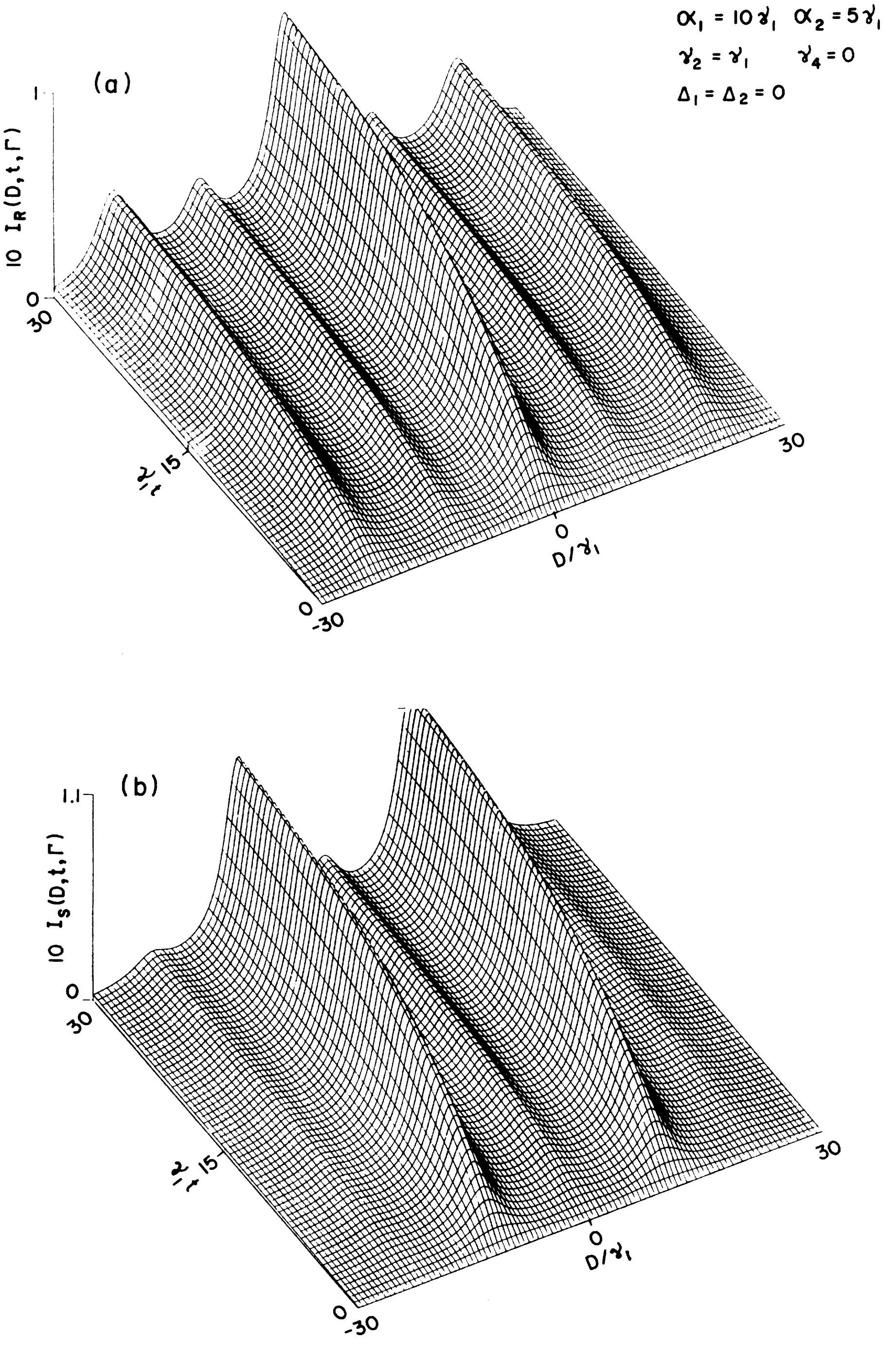

FIG. 3. Same as Fig. 2, but with $\gamma_3=1$.

with

$$\epsilon_1=\Gamma_{ij},\quad \delta_1=\nu,\quad \xi_1=\Omega_{ij},\quad \epsilon_2=\delta_3=\Gamma_{kl},\quad \epsilon_3=\Gamma_{kj}, \qquad (3.30)$$
$$\delta_2=\mu_1=\mu_2=0,\quad \xi_3=\Omega_{kj},\quad \mu_3=\xi_2=\Omega_{kl}.$$

In order to highlight the temporal development of the spectra we display in Figs. 2–4, the time-dependent Rayleigh and Stokes spectra with the atom initially in the ground state. Figure 2 shows the spectra for the resonance case in which the nonradiative damping terms γ_3 and γ_4 are zero. In this case it is known that there is no steady-state spectrum due to population trapping.[21] Figure 2 clearly shows the transient spectra which, however, vanish for large times. Note further that the spectra are symmetric about the center, which is characteristic of resonance excitation. A case where the spectra do not vanish in the steady state is shown in Fig. 3, $\gamma_3 \neq 0$ and $\gamma_4 = 0$. As expected, the spectra develop into symmetric five peaks as t increases. The time development of the off-resonance spectra shown in Fig. 4 exhibits further interesting behavior. Unlike in the resonance case, where the central peak and the sidebands maintain a consistent pattern while developing towards the steady state, the off-resonance spectra show some competitive behavior in the temporal development of the various peaks, as seen in Fig. 4.

In Fig. 5, we show the effects of the initial-state preparation of the atom on the development of the transient Rayleigh spectra with the other data the same as in Fig. 4. In Fig. 5(a), we assume the atom to be initially in the dressed state $|\psi_1\rangle$. For a small value of time ($\gamma_1 t = 0.1$), only the central peak and a right-hand sideband start developing. For longer times ($\gamma_1 t = 1$), more peaks begin to arise, and for sufficiently large time

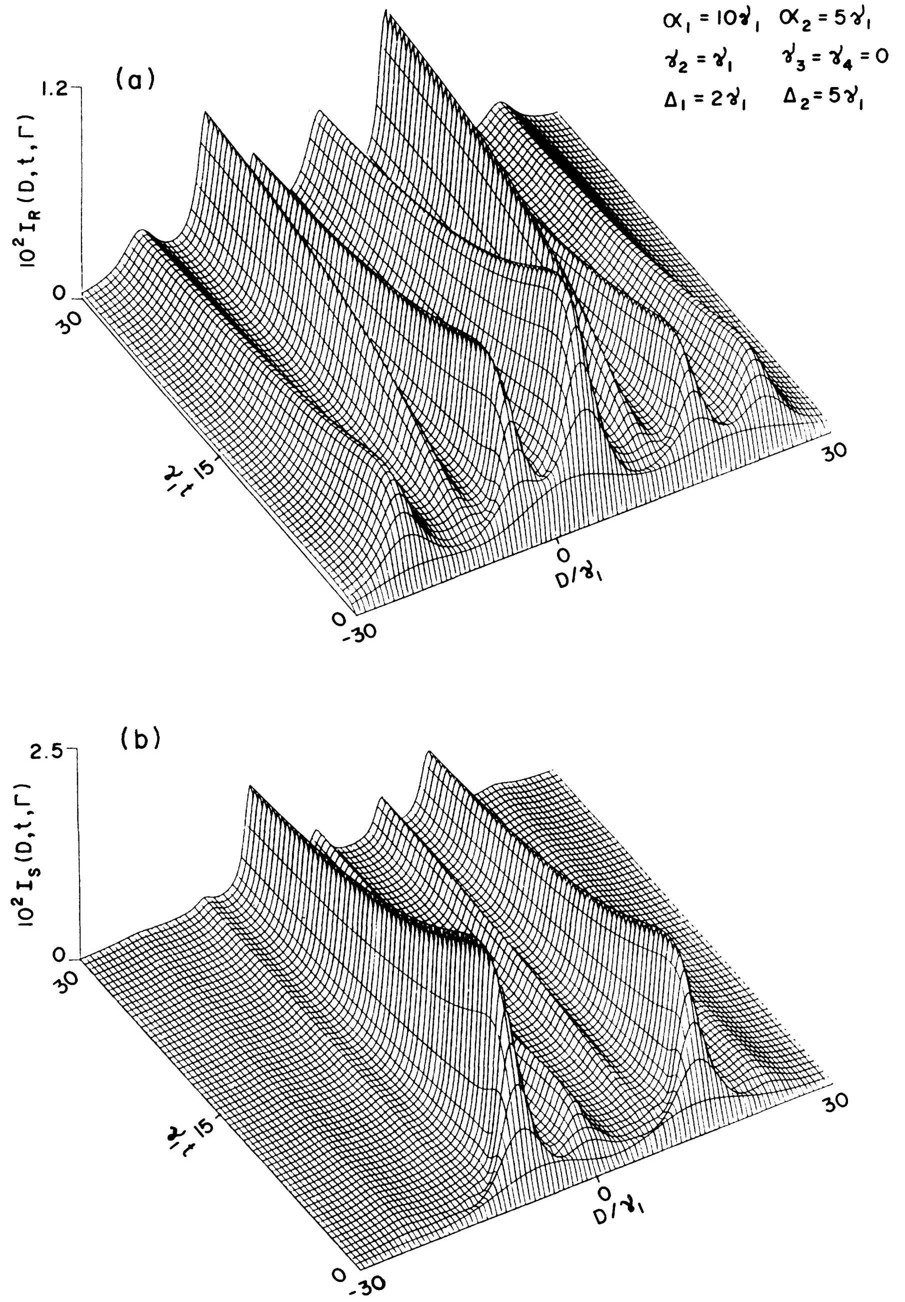

FIG. 4. Time development of (a) Rayleigh and (b) Stokes spectra when the driving fields are detuned with respect to the atomic transitions, with $\Gamma = 0.1$ and the atom prepared initially in the ground state.

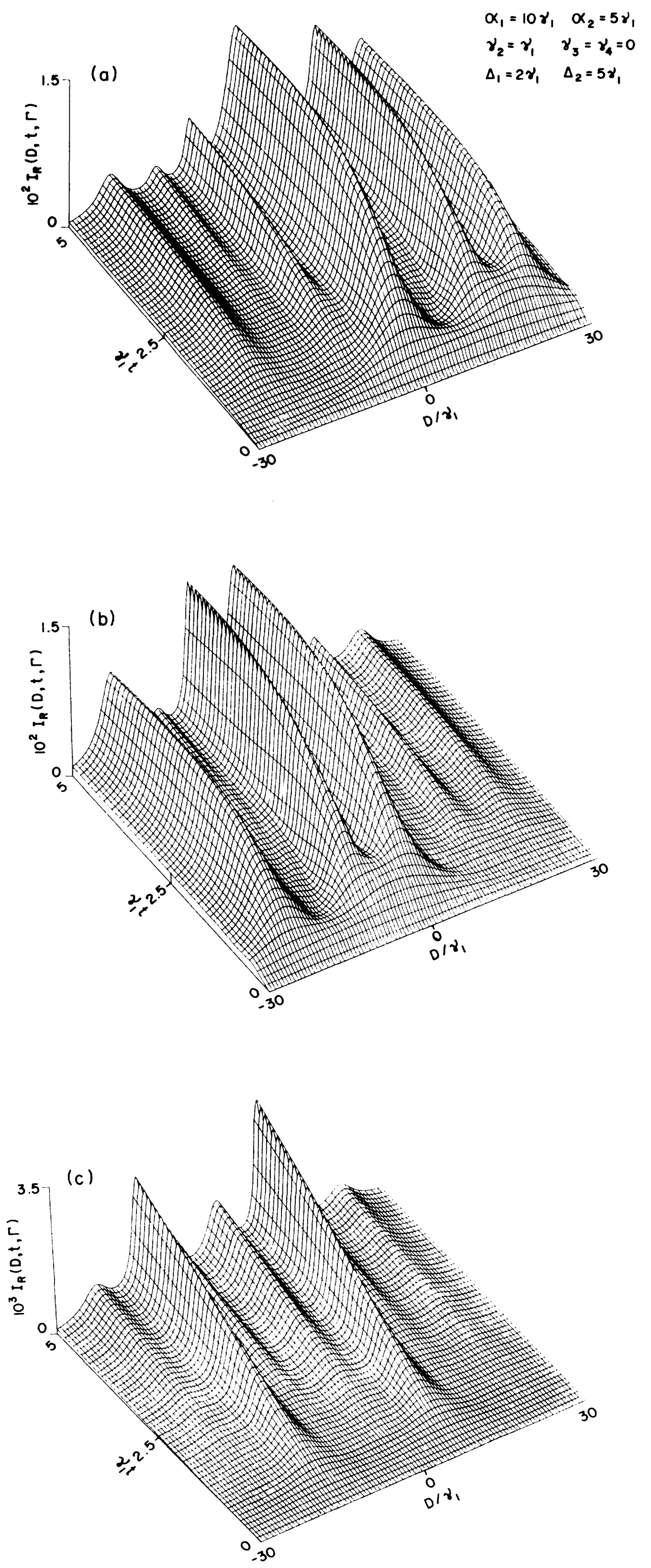

FIG. 5. Effect of initial state preparation of the atom on the development of the time-dependent Rayleigh spectrum. (a), (b), and (c) correspond to the atom initially in the dressed state $|\psi_1\rangle$, $|\psi_2\rangle$, and $|\psi_3\rangle$, respectively. Other data as in Fig. 4.

($\gamma_1 t = 5$) all seven peaks are clearly resolved. Figure 5(b), where the atom is assumed to be initially in the dressed state, $|\psi_2\rangle$ also shows a similar behavior, except that here the central peak and a left-hand sideband begin to develop initially. The case where the atom is initially in the dressed state $|\psi_3\rangle$ is even more interesting. This is illustrated by the curves in Fig. 5(c). Here the development of the transient spectrum begins nearly symmetrically with the appearance of the two near sidebands. Subsequently, for intermediate time ($\gamma_1 t = 2$), the central peak begins to appear, and for larger times ($\gamma_1 t = 5$), all the seven peaks are clearly resolved. In each of these three cases, the transient spectra ($\gamma_1 t \rightarrow \infty$) ultimately attain the same steady-state limit shown in Fig. 4.

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