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Two mechanisms for inversionless amplification in four-level atoms with Raman pumping

C. H. Keitel,* O. A. Kocharovskaya,† L. M. Narducci,‡ M. O. Scully,§ and S.-Y. Zhu‖
Max-Planck Institute für Quantenoptik, D-85740 Garching, Germany

H. M. Doss
Department of Physics, Mississippi State University, Mississippi State, Mississippi 39762
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We analyze the physical origin of the inversionless amplification in four-level atoms driven by a Raman field and identify the existence of two different mechanisms for the predicted gain. The first mechanism is connected with the appearance of a population inversion between the atomic states dressed by the coherent pump, while the second is linked to the buildup of coherence among these states. Proper selection of the relaxation parameters can make either the first or the second mechanism dominant over the other although usually both coexist.

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I. INTRODUCTION

Several possible schemes for inversionless amplification have been proposed during the past few years [1–11] for the purpose of reducing the ground-state absorption of an active medium by way of quantum interference effects among the various transitions. Recently two essentially different mechanisms have been identified [11–13]. The first, based on coherent population trapping, requires the transfer of some atoms into a special coherent superposition of lower levels which no longer interacts with a probe beam. The simplest realization of this scheme is provided by a three-level atom in a Λ configuration with the active medium prepared in a nonabsorbing state (see, for example, Refs. [1,3] and Fig. 1 of Ref. [11]) and enough population in the upper laser level to support an inversion with the optically active lower dressed state.

The second mechanism is linked to the existence of interference effects among dressed states. In the p and h configurations (see Fig. 1 of Ref. [11]), where this mechanism is active in its simplest form, the amplification process can be interpreted as a form of anti-Stokes Raman scattering of the coherent pump field. In this case the existence of gain requires the absence of a population inversion at the forbidden transition, a constraint which is rather unusual for this type of atomic configuration. The second mechanism is operative also when two upper atomic sublevels decay to the same continuum [2,4,13,14] or when two lower levels are pumped by an incoherent field [15], although in this case the atomic coherence does not emerge from the action of an external driving field.

These schemes can be regarded as prototype examples of active media where either the first or the second mechanism for lasing without inversion can occur without competition from the other. Both mechanisms, however, can coexist, as shown recently by one of us [14], if a three-level active medium can support the amplification of a bichromatic field when driven by a pump which is resonant with the low-frequency transition.

Previous investigations of four-level atoms with Raman pumping have emphasized the existence of noninversion lasing by way of the first mechanism [10,16,17]. Thus this form of coherent pumping is suitable not only for the creation of the required coherent superposition states, but it also leads to gain as a result of a population inversion between dressed states, at least for selected ranges of atomic parameters [16,17]. Furthermore, the interference among the dressed states generally hinders the amplification process. Hence it follows that the dressed-states inversion must be large enough to overcome the absorptive effect of the interference contribution. The full range of parameters, and in particular the possibility of rapid relaxation between the two closely spaced lower levels, has not been investigated in sufficient detail; as it turns out, by extending the analysis of this model to a broader range of parameters, we find that the second mechanism can also become effective and can coexist with the first.

The purpose of this paper is to show that four-level atoms with a Raman pump can support both mechanisms, in general. In addition, we wish to define the conditions under which either the first or the second process dominates. In particular, we show that the interference among dressed states can yield not only a positive contribution to the gain, but it can also lead to amplification even in the absence of dressed-state population inversion.

In Sec. II we analyze this problem in its most natural setting of the dressed atomic states; we derive a simple formula of the absorption spectrum of a weak probe and identify the dressed population and coherence contributions to the amplification process. In Sec. III we discuss the same problem in terms of bare states. From this perspective, gain can only be the consequence of atomic coherence, of course. We show, however, that the two mechanisms of amplification can be linked to specific bare polarization components, each of which may become the sole source of gain under appropriate conditions.

II. DRESSED-STATE ANALYSIS OF THE TWO AMPLIFICATION MECHANISMS

We are interested in the behavior of the absorption or gain spectrum [18] of a weak probe field of tunable frequency ω that propagates through an active medium of four-level atoms whose energy-level diagram is shown schematically in Fig. 1 [16]. The atoms are driven coherently by a classical Raman field with carrier frequency $\omega_R=\frac{1}{2}(\omega_{42}+\omega_{41})$ and are described by the semiclassical Hamiltonian

$$H=H_0+H_1 \ , \tag{1}$$

where

$$H_0=\hbar\omega a_3^\dagger a_3+\hbar\omega_R a_4^\dagger a_4 \tag{2}$$

and

$$\begin{aligned} H_1=&-\hbar\Delta a_1^\dagger a_1+\hbar\Delta a_2^\dagger a_2-\hbar\delta a_3^\dagger a_3 \\ &+\hbar g_R[\exp(-i\omega_R t)(a_4^\dagger a_1+a_4^\dagger a_2)+\text{H.a.}] \\ &+\hbar g[\exp(-i\omega t)(a_3^\dagger a_1+a_3^\dagger a_2)+\text{H.a.}] \ . \end{aligned} \tag{3}$$

The operators $a_i^\dagger$ and a_i are the usual Fermion creation and annihilation operators for electrons, g_R and g are the Rabi frequencies of the Raman and probe fields, respectively, and Δ is half of the frequency spacing between the two lowest levels ($\Delta=\frac{1}{2}\omega_{21}$); $\hbar\omega_R$ denotes the energy of level 4 measured from the midpoint of the ground-state doublet, and $\delta\equiv\omega-\omega_3$ is the detuning of the probe field from the frequency $\omega_3=\frac{1}{2}(\omega_{31}+\omega_{32})$. This model includes an incoherent pump mechanism to transfer ground-state atoms to the upper lasing level at the rate R. As shown in Refs. [10,16], a minimum, nonzero, incoherent pump rate is always required to produce inversionless gain for selected values of the remaining parameters, and especially the strength of the Raman field.

In Ref. [16] we calculated the absorption spectrum using the formalism of the linear-response theory; here we derive the same quantity by the equivalent process based on the linearization of the Maxwell-Bloch equations with respect to the strength of the probe field. Because this field interacts only with the $3\rightarrow1$ and $3\rightarrow2$ transitions, its absorption coefficient is given by the sum of the imaginary parts of the off-diagonal matrix elements ρ_{31} and ρ_{32} of the atomic density operator. As expected, the results of these two approaches are identical.

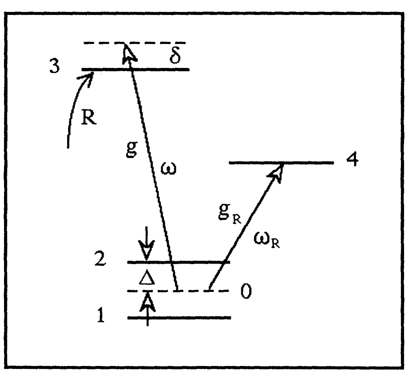

FIG. 1. Schematic representation of the four-level atom driven by a Raman field with Rabi frequency g_R and carrier frequency ω_R. Δ is one-half of the frequency spacing ω_{21} between levels 1 and 2; g and ω denote, respectively, the Rabi and carrier frequencies of the probe field; R is the incoherent pump rate from the ground-state doublet to level 3; and δ is the detuning between the probe field frequency and ω_3, measured from the chosen origin of the energy axis.

As the starting point of this calculation we transform the Hamiltonian (3) into the interaction picture and project it on the basis of the dressed states $|s\rangle$, $|r\rangle$, and $|t\rangle$ of the driven atom. These are the eigenstates of the interaction Hamiltonian without the weak probe field contribution and their explicit form in terms of bare states is [16]

$$|s\rangle=\sin^2\frac{\theta}{2}|1\rangle+\cos^2\frac{\theta}{2}|2\rangle+\frac{1}{\sqrt{2}}\sin\theta|4\rangle \ , \tag{4a}$$

$$|r\rangle=\frac{1}{\sqrt{2}}\sin\theta(|1\rangle-|2\rangle)+\cos\theta|4\rangle \ , \tag{4b}$$

$$|t\rangle=-\cos^2\frac{\theta}{2}|1\rangle-\sin^2\frac{\theta}{2}|2\rangle+\frac{1}{\sqrt{2}}\sin\theta|4\rangle \ , \tag{4c}$$

where

$$\sin\theta=\frac{\sqrt{2}g_R}{G} \ , \quad \cos\theta=\frac{\Delta}{G} \ , \quad G=\sqrt{\Delta^2+2g_R^2} \ . \tag{5}$$

After this transformation the interaction Hamiltonian takes the form

$$\begin{aligned} \tilde{H}_1=&-\hbar\delta a_3^\dagger a_3+\hbar G|s\rangle\langle s|-\hbar G|t\rangle\langle t| \\ &+\hbar g[|3\rangle\langle s|-|3\rangle\langle t|+\text{H.a.}] \ , \end{aligned} \tag{6}$$

where the tilde denotes the interaction picture.

The next step is the derivation of the appropriate Maxwell-Bloch equations for the driven atom plus probe system in the dressed- state representation, with the inclusion of the phenomenological incoherent decay and pump terms. The interaction picture equations for the off-diagonal matrix elements of the atomic density operator which are needed for the construction of the absorption spectrum are listed Appendix A for completeness.

These equations can be solved analytically in steady state and, to within first order in the probe field strength, lead to the following expression for the absorption spectrum $A(\delta)$:

$$A(\delta)=\frac{1}{g}\text{Im}(\rho_{31}+\rho_{32})=\frac{1}{g}\text{Im}(\rho_{t3}-\rho_{s3})\equiv A_P+A_C \ , \tag{7}$$

where

$$\begin{aligned} A_P(\delta)=&\text{Re}(M_{11}-M_{31})(\rho_{33}-\rho_{ss}) \\ &+\text{Re}(M_{33}-M_{13})(\rho_{33}-\rho_{tt}) \ , \end{aligned} \tag{8a}$$

represents the contribution of the dressed-states populations and

$$\begin{aligned} A_C(\delta)=&\text{Re}[(M_{33}-M_{13})\rho_{ts}+(M_{11}-M_{31})\rho_{st} \\ &+(M_{12}-M_{32})(\rho_{rt}-\rho_{rs})] \end{aligned} \tag{8b}$$

represents the corresponding contribution of the dressed atomic coherences (off-diagonal matrix elements of the density operator). For convenience we list the explicit expressions for the matrix elements M_{ij} in Appendix B. The density-matrix elements that appear in Eqs. (7) and (8) denote their steady-state values; those listed in Eq. (7) are calculated to within first order in the probe field strength and those in Eq. (8) to zeroth order. As expected, this procedure yields exactly the same result as derived in Ref. [16] [see Eqs. (17a) and (17b) of Ref. [16]] following the regression theorem approach.

The spectral contributions $A_P(\delta)$ and $A_C(\delta)$, given by Eqs. (8a) and (8b), originate from two fundamentally different physical processes and their dependence upon the probe frequency is illustrated in Figs. 2 and 3 for different sets of parameters. In Fig. 2 the coherence decay rate γ_{12} of the two lower levels is smaller than W_4, the spontaneous decay rate of level 4 to the ground states 1 and 2, and consequently also smaller than the coherence decay rate γ_{34} of the two upper levels. In this case, gain without inversion between the bare states is mainly the consequence of an inversion between the dressed-state populations (we refer to this process as the first mechanism). In Fig. 3, instead, we select parameters that correspond to the opposite limit, i.e., a slower decay of the coherence of the two upper levels relative to that of the two lower states; in this case gain arises from the coherence contribution A_C (second mechanism).

We can acquire a better insight into the physical origin of these two mechanisms by deriving a simpler analytical expression for the absorption spectrum [Eq. (7)] in the limit in which the collisional decay rate W_{34} of the dipole forbidden transition between the upper levels and the fre-

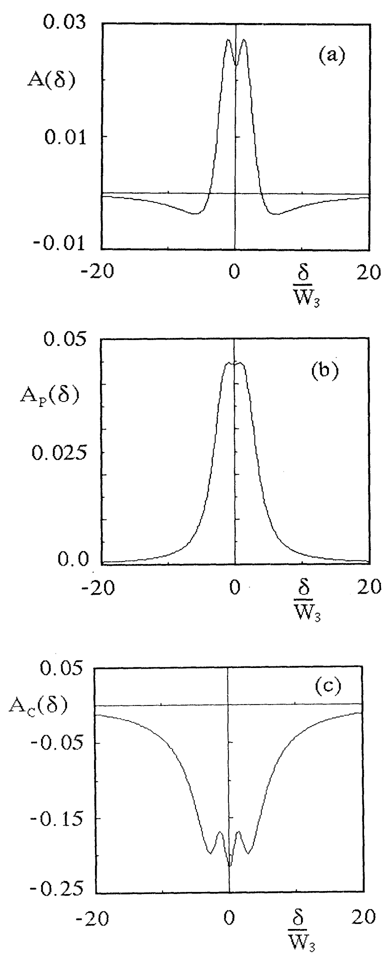

FIG. 2. (a) Gain spectrum of the probe field as a function of the scaled detuning δ/W_3. The parameters of the simulation are $R=0.6$, $W_3=1.0$, $W_4=2.5$, $W_2=W_{34}=0.1$, $\Delta=0.5$, and $g_R=1.5$. The stationary values of the population density-matrix elements are $\rho_{11}=\rho_{22}=0.3566$, $\rho_{33}=0.2038$, $\rho_{44}=0.0830$; the corresponding stationary dressed matrix elements are $\rho_{ss}=\rho_{tt}=0.1407$, $\rho_{rr}=0.5147$, $\rho_{33}=0.2038$. (b) Population contribution to the gain spectrum [Eq. (8a)]. (c) Coherence contribution to the gain spectrum [Eq. (8b)].

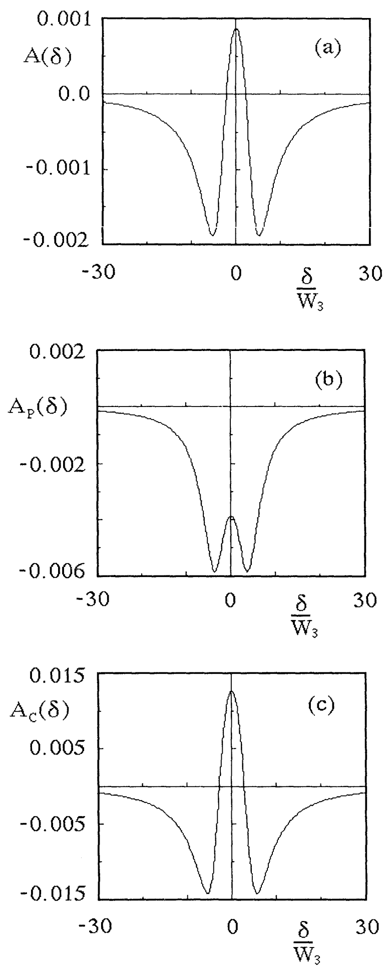

FIG. 3. (a) Gain spectrum of the probe field as a function of the scaled detuning δ/W_3. The parameters of the simulation are $R=0.7$, $W_3=1.0$, $W_4=1.5$, $W_2=5.0$, $W_{34}=0.1$, $\Delta=0.5$, and $g_R=2.5$. The stationary values of the population density-matrix elements are $\rho_{11}=\rho_{22}=0.3087$, $\rho_{33}=0.2058$, $\rho_{44}=0.1767$; the corresponding stationary dressed matrix elements are $\rho_{ss}=\rho_{tt}=0.2230$, $\rho_{rr}=0.3481$, $\rho_{33}=0.2058$. (b) Population contribution to the gain spectrum [Eq. (8a)]. (c) Coherence contribution to the gain spectrum [Eq. (8b)].

quency spacing 2Δ between the two lower levels are sufficiently small, in comparison with the remaining rates, to be negligible. This constraint on W_{34} is not too stringent, at least for sufficiently low gas pressure, because of the forbidden nature of the $3\rightarrow 4$ relaxation process. In a real system, instead, Δ may not be as small as required for the validity of our approximate formulas. We must stress, however, that even if Eqs. (9a) and (9b) do not apply, the physical classification of the two mechanisms still holds, as shown in Figs. 2 and 3, which we constructed using the exact equations. What is lost in this case is the ability to provide a transparent physical interpretation.

When this constraint on W_{34} and Δ holds, Eqs. (8a) and (8b) take the explicit form

$$A_P(\delta)=\frac{2}{N|D|^2}[\gamma_{34}(2g_R^2+\gamma_3\gamma_{34})+\gamma_3\delta^2] \times\left[\frac{R}{W_3}\left[\frac{\gamma_4 W_4}{2g_R^2}+1+\frac{W_4}{\gamma_{12}}\right]-\left[1+\frac{W_4\gamma_4}{4g_R^2}\right]\right] \tag{9a}$$

and

$$A_C(\delta)=\frac{2W_4}{N|D|^2}\left[(2g_R^2+\gamma_3\gamma_{34})\left[1-\frac{\gamma_{34}\gamma_4}{4g_R^2}\right]-\delta^2\left[1+\frac{\gamma_3\gamma_4}{4g_R^2}\right]\right], \tag{9b}$$

where

$$N=\left[2+\frac{R}{W_3}\right]\left[\frac{\gamma_4 W_4}{2g_R^2}+1+\frac{W_4}{\gamma_{12}}\right]+1\ , \tag{10a}$$

$$|D|^2=(2g_R^2-\delta^2+\gamma_3\gamma_{34})^2+(\gamma_3+\gamma_{34})^2\delta^2\ . \tag{10b}$$

The symbol γ_3 denotes the common decay rate of the polarizations ρ_{31} and ρ_{32}, and γ_4 the corresponding rate for ρ_{41} and ρ_{42}; W_3 is the common spontaneous decay rate of the $3\rightarrow 1$ and $3\rightarrow 2$ transitions, and R is the incoherent pump rate to level 3. A sketch of the derivation leading to Eqs. (9) is given in Appendix C.

The nature of the two gain mechanisms and the role of the dressed-state populations and polarizations contribution can be best understood if we consider the absorption spectrum in the resonant limit ($\delta=0$) where the net absorption $A=A_P+A_C$ is proportional to

$$A\propto\frac{R}{W_3}\left[\frac{W_4\gamma_4}{2g_R^2}+1+\frac{W_4}{\gamma_{12}}\right]-\left[1+\frac{W_4\gamma_4}{4g_R^2}\right]+\frac{W_4}{\gamma_{34}}\left[1-\frac{\gamma_{34}\gamma_4}{4g_R^2}\right]. \tag{11}$$

The first two terms on the right-hand side of Eq. (11) correspond to the dressed population contribution A_P, while the last term is related to the dressed coherence A_C. The term proportional to the pump rate R describes the effect of the stimulated emission process from the upper dressed-state level, while the second term measures the absorption contribution of the lower dressed-state populations.

From Eq. (11) we see that, in the limit $\gamma_{12}\ll W_4$ (long-lived coherence of the lowest levels), and if g_R is not much smaller than W_4, γ_4, and γ_{34}, the stimulated emission contribution from the upper dressed state dominates both the absorptive and coherence contributions. This continues to be true even in the case when $R<W_3$, i.e., when there is no population inversion between the bare-state lasing levels. In addition, if $\gamma_{12}>4g_R^2/\gamma_4$ (long-lived coherence of the upper two levels), the dressed coherence contributes no gain, and the first mechanism of lasing without inversion can be observed in its pure form. If, one the other hand, the Raman field amplitude satisfies the inequality $4g_R^2>\gamma_{34}\gamma_4$, the dressed coherence can provide a contribution to the laser gain; if, in addition, the pump parameter R is sufficiently small, the laser gain originates exclusively from the dressed coherence contribution (second mechanism).

In the next section we investigate the origin of the laser gain in the bare-state representation and show that the above two mechanisms for inversionless amplification can also be interpreted as emerging from two distinct polarization components of the active medium.

III. BARE-STATE ANALYSES OF THE GAIN

In the preceding section we have shown that gain without inversion between the bare states of an active medium can arise from a population inversion between dressed states and/or from dressed coherencies, depending on the selection of the decay parameters. In this section we show that these two different modes of inversionless laser action can also be interpreted as the result of gain induced by two different types of bare-state coherencies.

For this purpose, it is useful to recall the equation of motion for the density-matrix element ρ_{31}:

$$\frac{d\rho_{31}}{dt}=-(\gamma_3+i\omega_{31})\rho_{31}+i[g(\rho_{33}-\rho_{11}-\rho_{21})+g_R\rho_{34}]\ , \tag{12}$$

whose stationary behavior, together with that of ρ_{32}, controls the small signal probe gain and absorption in the frequency range of interest. Unlike the corresponding equation for a traditional two-level laser model, the right-hand side of Eq. (12) contains two off-diagonal elements ρ_{21} and ρ_{34} that can have a very significant effect on the usual gain requirement (i.e., $\rho_{33}>\rho_{11}$). The magnitude of these off-diagonal elements is a measure of the induced coherence between the indicated atomic levels and their importance in producing amplification or absorption of a weak probe.

At the end of the preceding section we concluded that the first amplification mechanism (dressed population inversion) dominates if the decay rate γ_{12} is much smaller than γ_{34}, i.e., the rate at which levels 3 and 4 lose memory of their correlation. This observation suggests that the first mechanism is closely related to the presence of a strong lower level coherence ρ_{12} whose magnitude in steady state depends on the smallness of its decay rate.

We also noted that the second amplification mechanism (gain due to dressed coherencies) acquires a dominant role if the decay rate γ_{34} of the upper levels coherence is of the order of, or preferably smaller than, the Rabi frequency of the Raman driving field. From the structure of the right-hand side of Eq. (12) it is plausible to assume that the second mechanism is associated with the emergence of a strong coherent link between levels 3 and 4. In the remainder of this section we provide a more formal support for these intuitive statements.

For this purpose we consider the absorption spectra given by Eq. (8). We replace the dressed density-matrix elements (e.g., ρ_{ss}, ρ_{rt}, etc.) by their expressions in terms of bare states [see eqs. (C5) in Appendix C], we substitute the calculated values of M_{ij} in the limiting case W_{34}, $\Delta\to 0$ [see Appendix B], and, for simplicity, we consider the case of a resonant probe ($\delta=0$). The result of this simple calculation is

$$A_P(\delta=0)=\frac{2\gamma_{34}}{\gamma_3\gamma_{34}+2g_R^2}[\rho_{33}-\tfrac{1}{2}(\rho_{11}+\rho_{12}+\rho_{44})]\ , \tag{13a}$$

$$A_C(\delta=0)=\frac{2}{\gamma_3\gamma_{34}+2g_R^2}[\tfrac{1}{2}\gamma_{34}(\rho_{44}-\rho_{11}-\rho_{12})+W_4\rho_{44}]\ . \tag{13b}$$

The total absorption coefficient, in resonance, is the sum of the two contribution (13a) and (13b), i.e.,

$$A(\delta=0)=\frac{2}{\gamma_3\gamma_{34}+2g_R^2}[(\rho_{33}-\rho_{11}-\rho_{12})\gamma_{34}+W_4\rho_{44}]\ . \tag{14}$$

This expression for the small signal absorption coefficients is to be compared with the stationary expressions for the matrix elements ρ_{31} [Eq. (12)] and ρ_{32}, which together acts as the source terms of Maxwell's equation for the probe field. The off diagonal element ρ_{12} of the atomic density operator in Eq. (14) originates from both the dressed-state populations of the lower levels [Eq. (13a)] and from the real part of the dressed coherence ρ_{st} [Eq. (13b)], as shown explicitly by Eq. (C5d). The term $W_4\rho_{44}$, which in steady state is proportional to $\mathrm{Im}\rho_{41}$, is connected to the imaginary part of ρ_{st}. Thus a comparison of Eqs. (12) and (14) shows that the bare-state coherence ρ_{34} is linked only to the dressed coherence and hence is responsible for the second mechanism of inversionless gain. In addition, the real part of the dressed coherence is negligible when either of the two mechanisms acquires a dominant role, so that ρ_{12} in this case is connected primarily to the dressed populations part of the gain, or to the first mechanism.

Hence we conclude that the first gain mechanism is the consequence of the ρ_{12} coherence, while the second is connected to the appearance of a strong correlation between the upper bare states 3 and 4 by way of the ρ_{34} coherence. Because ρ_{34} is dynamically coupled to the off-diagonal elements ρ_{31} and ρ_{32} by a process which is reminiscent of an anti-Stokes transition mediated by the external driving field, it is not surprising that this mechanism of lasing without inversion should be viewed as being fundamentally different from the one that originates from the ρ_{12} coherence.

APPENDIX A

The relevant off-diagonal elements of the dressed density operator satisfy the vector differential equation

$$\frac{d\boldsymbol{\psi}}{dt}=L\boldsymbol{\psi}+\boldsymbol{\phi}\ , \tag{A1}$$

where

$$L=\begin{bmatrix} -i(G+\delta)-\alpha_1 & \beta_1 & \beta_2 \\ \beta_1 & -i\delta-\alpha_2 & \beta_1 \\ \beta_2 & \beta_1 & i(G-\delta)-\alpha_1 \end{bmatrix}\ , \tag{A2}$$

$$\boldsymbol{\psi}=\begin{bmatrix}\rho_{s3}\\ \rho_{r3}\\ \rho_{t3}\end{bmatrix}\ ,\quad \boldsymbol{\phi}=\begin{bmatrix}-ig(\rho_{33}-\rho_{ss}-\rho_{st})\\ ig(\rho_{rs}-\rho_{rt})\\ ig(\rho_{33}-\rho_{tt}+\rho_{ts})\end{bmatrix}\ , \tag{A3}$$

and

$$\alpha_1=\gamma_3\frac{1+\cos^2\theta}{2}+\gamma_{34}\tfrac{1}{2}\sin^2\theta\ , \tag{A4a}$$

$$\alpha_2=\gamma_3\sin^2\theta+\gamma_{34}\cos^2\theta\ , \tag{A4b}$$

$$\beta_1=(\gamma_3-\gamma_{34})\frac{1}{\sqrt{2}}\sin\theta\cos\theta\ , \tag{A4c}$$

$$\beta_2=(\gamma_3-\gamma_{34})\tfrac{1}{2}\sin^2\theta\ . \tag{A4d}$$

The symbol γ_3 denotes the common relaxation rate of the ρ_{31} and ρ_{32} polarization components, and γ_{34} is the corresponding rate of decay of ρ_{34}.

APPENDIX B

The matrix M is defined as $M=(-L)^{-1}$, with L given by Eq. (A2). Its matrix elements are

$$M_{ij}=\frac{1}{\det(-L)}\mathrm{cof}_{ij}\ , \tag{B1}$$

where cof denotes cofactor,

$$\mathrm{cof}_{11}=(i\delta+\alpha_2)(i\delta-iG+\alpha_1)-\beta_1^2\ , \tag{B2a}$$

$$\mathrm{cof}_{12}=\mathrm{cof}_{21}=\beta_1(i\delta-iG+\alpha_1+\beta_2)\ , \tag{B2b}$$

$$\mathrm{cof}_{13}=\mathrm{cof}_{31}=\beta_1^2+\beta_2(i\delta+\alpha_2)\ , \tag{B2c}$$

$$\mathrm{cof}_{22}=(i\delta+\alpha_1)^2+G^2-\beta_2^2\ , \tag{B2d}$$

$$\mathrm{cof}_{23}=\mathrm{cof}_{32}=\beta_1(i\delta+iG+\alpha_1+\beta_2)\ , \tag{B2e}$$

$$\mathrm{cof}_{33}=(i\delta+\alpha_2)(i\delta+iG+\alpha_1)-\beta_1^2\ , \tag{B2f}$$

and

$$\det(-L)=(i\delta+\alpha_2)[(i\delta+\alpha_1)^2+G^2]-2\beta_1^2(i\delta+\alpha_1+\beta_2)-\beta_2^2(i\delta+\alpha_2)\ . \tag{B3}$$

APPENDIX C

The purpose of this appendix is to outline the derivation of an explicit expression for the absorption spectrum $A(\delta)$ in the limit in which both W_{32} and Δ are negligibly small in comparison with the remaining rates of the problem. We begin by listing the equations of motion for the atomic matrix elements ρ_{ij} to lowest order to the probe field strength. These equations are

$$\frac{d\rho_{11}}{dt}=\frac{d\rho_{22}}{dt}=W_3\rho_{33}+W_4\rho_{44}-R\rho_{11}+W_2(\rho_{22}-\rho_{11})+2g_R\,\mathrm{Im}\rho_{41}\ , \tag{C1a}$$

$$\frac{d\rho_{44}}{dt}=-2W_4\rho_{44}-4g_R\,\mathrm{Im}\rho_{41}\ , \tag{C1b}$$

$$\frac{d\rho_{33}}{dt}=2R\rho_{11}-2W_3\rho_{33}\ , \tag{C1c}$$

$$\frac{d\rho_{12}}{dt}=-\gamma_{12}\rho_{12}+2g_R\,\mathrm{Im}\rho_{41}\ , \tag{C1d}$$

$$\frac{d}{dt}\mathrm{Im}\rho_{41}=-(W_4+\tfrac{1}{2}\gamma_{12})\mathrm{Im}\rho_{41}+g_R(\rho_{44}-\rho_{11}-\rho_{12})\ , \tag{C1e}$$

where

$$\gamma_{ij}=\tfrac{1}{2}\sum_k (W_{ik}+W_{jk})\ , \tag{C2}$$

and $R=W_{13}=W_{23}$.

The steady-state solutions of Eq. (C1) can be written in the form

$$\rho_{11}=\rho_{22}=\frac{\sigma}{N}\ , \tag{C3a}$$

$$\rho_{44}=\frac{1}{N}\ , \tag{C3b}$$

$$\rho_{33}=\frac{\lambda\sigma}{N}\ , \tag{C3c}$$

$$\rho_{12}=-\frac{1}{\mu N}\ , \tag{C3d}$$

$$\mathrm{Im}\rho_{41}=-\frac{1}{2}\frac{\nu}{\mu N}\quad(\mathrm{Re}\rho_{41}=0)\ , \tag{C3e}$$

where we have introduced the symbols

$$N=(2+\lambda)\sigma+1\ , \tag{C4a}$$

$$\sigma=\left[\frac{\nu}{\mu}+\frac{\nu}{2}\right]\frac{\nu}{2\mu}+1+\frac{1}{\mu}\ , \tag{C4b}$$

$$\lambda=\frac{R}{W_3}\ ,\quad \mu=\frac{\gamma_{12}}{W_4}\ ,\quad \nu=\frac{\gamma_{12}}{g_R}\ . \tag{C4c}$$

The corresponding zeroth-order expressions for the dressed density-matrix elements are

$$\rho_{ss}=\rho_{tt}=\tfrac{1}{2}(\rho_{11}+\rho_{12}+\rho_{44})=\frac{1}{2}\frac{\sigma+1-\frac{1}{\mu}}{N}\ , \tag{C5a}$$

$$\rho_{rr}=\rho_{11}-\rho_{12}=\frac{\sigma+\frac{1}{\mu}}{N}\ , \tag{C5b}$$

$$\rho_{rs}=\rho_{rt}=0\ , \tag{C5c}$$

$$\rho_{st}=\tfrac{1}{2}(\rho_{44}-\rho_{11}-\rho_{12})-\sqrt{2}i\,\mathrm{Im}\rho_{41}=\frac{1}{N}\frac{\nu}{\mu}\left[-\frac{\nu}{4}\left[\frac{1}{\mu}+\frac{1}{2}\right]+\frac{i}{\sqrt{2}}\right]\ . \tag{C5d}$$

Finally, with the help of the matrix elements M_{ij} listed in Appendix B, we arrive at Eqs. (9).

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*Permanent address: Imperial College, Blackett Laboratory, Optics Section, London SW7 2BZ, United Kingdom.

†Permanent address: Institute of Applied Physics, Russian Academy of Science, 603600 Nizhny Novgorod, Russia.

‡Permanent address: Department of Physics and Atmospheric Sciences, Drexel University, Philadelphia, PA 19104.

§Also at Department of Physics, Texas A&M University, College Station, TX 77843-4242.

‖Permanent address: Department of Applied Physics, Shanghai Jiao Tong University, Shanghai 200030, People's Republic of China.

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[18] In the following discussion we shall use "absorption" or "gain" spectrum interchangeably, as we find appropriate, in view of the fact that even for the same parameters one can produce attenuation or amplification of a probe beam by simply varying its frequency.