Large population recovery in a strongly driven two-level atomic system by an additional laser field

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In a saturated two-level atomic system driven by a strong, near-resonant field, a large population difference between the two energy levels is recovered by an additional strong off-resonant laser field. This implies that the stimulated transition between the energy levels is inhibited by the additional laser field. This effect is attributed to the transverse resonance which has been extensively studied in the radio-frequency spectroscopy. It is also shown that the definite dispersion-free condition is realized when the intensities of the saturating field and the second field are the same.

I. INTRODUCTION

The interaction of two waves in a nonlinear medium is one of the basic problems of the theory of multimode-laser and laser spectroscopy.[1,2] In this paper, we discuss the recovery of the population difference by an additional (second) laser field in the two-level system saturated by an intense (saturating) field. This implies that the saturated transition between the two energy levels by the saturating-laser field can be inhibited by the second-laser field. This effect can be explained in terms of the transverse resonance[3] in rf spectroscopy.

Let us consider an atom with two energy eigenstates $|a\rangle$ and $|b\rangle$ which interact with the two-laser fields,

$$E(t)=\tfrac{1}{2}[E_1\exp(-i\omega_1 t)+E_2\exp(-i\omega_2 t)+\text{c.c.}]\ , \tag{1}$$

where ω_1 and ω_2 are the frequencies of the two-laser fields, and E_1 and E_2 are their field strengths. The behavior of a weak probe-wave absorption profile in the presence of a strong pump wave tuned directly to the resonance frequency has been studied theoretically by Mollow,[4] and experimentally in an atomic beam by Wu *et al.*[5] The dynamics of the two-level system interacting with two arbitrary-strength fields have been analyzed by Bonch-Bruevich *et al.*,[6] Tsukada,[7] Toptygina and Fradkin,[8] and by Agrawal and Nayak.[9]

According to a previous paper (Ref. 7), we study the static component of the population difference $d_0=(\rho_{aa}-\rho_{bb})^{dc}$ together with the absorption $\mathrm{Im}\rho_{ab}(\omega_2)$ and the dispersion spectra $\mathrm{Re}\rho_{ab}(\omega_2)$. For the convenience of the latter discussions, we introduce the following quantities:

$$\Omega_1=\mu_{ab}E_1/\hbar,\quad \Omega_2=\mu_{ab}E_2/\hbar\ , \tag{2}$$

where Ω_1 and Ω_2 are the field amplitudes denoted by the "Rabi frequency," and μ_{ab} is the atomic-dipole moment between the $|a\rangle$ and $|b\rangle$ states. The calculations were performed for the particular case with one decay rate $\gamma_1=\gamma_2=\gamma$ throughout this paper, where γ_1 and γ_2 are the longitudinal and transverse relaxation rates, and with $d_0^{(0)}=\rho_{aa}^{(0)}-\rho_{bb}^{(0)}=1$, where $\rho_{aa}^{(0)}$ and $\rho_{bb}^{(0)}$ are the mean thermal occupation numbers for states $|a\rangle$ and $|b\rangle$, respectively.

II. CALCULATION OF THE POPULATION-INVERSION SPECTRUM

In Fig. 1, we show the population difference d_0 as a function of $(\omega_2-\omega_1)/\gamma$ for the different values of the saturating field detuning $(\omega_0-\omega_1)/\gamma$, and for $\Omega_1=20\gamma$ and $\Omega_2=5\gamma$. The corresponding absorption function $\mathrm{Im}\rho_{ab}(\omega_2)$ and the dispersion function $\mathrm{Re}\rho_{ab}(\omega_2)$ have been reported in a previous paper.[10] We can see in Fig. 1 that there are two dominant resonant structures in each curve. A striking feature is that the resonance structures appear as an increase of the population difference d_0 when the saturating field is exactly on resonance $(\omega_1=\omega_0)$. Before discussing this peculiar character, we consider the positions of the dominant resonances.

In a frame rotating at the saturating-laser frequency

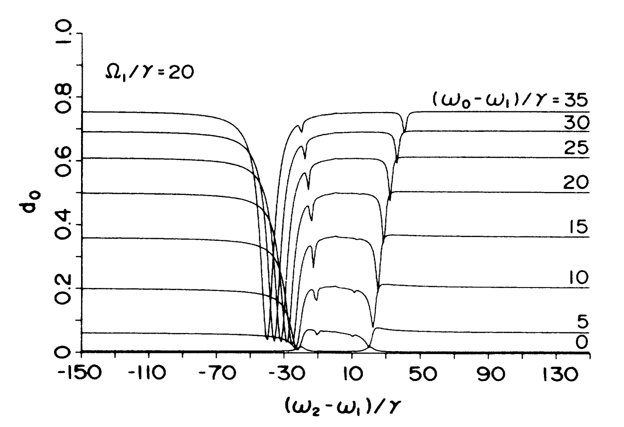

FIG. 1. Time-averaged population difference $d_0=(\rho_{aa}-\rho_{bb})^{dc}$ as a function of the second-field detuning $(\omega_2-\omega_1)/\gamma$ for the different values of the saturating-field detuning $(\omega_0-\omega_1)/\gamma$, and with constant-field strengths ($\Omega_1/\gamma=20$ and $\Omega_2/\gamma=5$).

ω_1, the atomic-state vector $\boldsymbol{\rho}$ obeys the well-known equation,[11,12] neglecting the decay term,

$$\frac{d\boldsymbol{\rho}}{dt}=\boldsymbol{\Omega}\times\mathbf{p}\ , \tag{3}$$

where $\boldsymbol{\Omega}$, the driving-field vector, is given by

$$\boldsymbol{\Omega}=(\Omega_1+\Omega_2\cos(\Delta\omega_{12}t),\Omega_2\sin(\Delta\omega_{12}t),\Delta_1)\ , \tag{4}$$

where $\Delta_1=\omega_0-\omega_1$ is the detuning of the saturating field from the unperturbed atomic-resonance frequency ω_0. The atomic-state vector precesses around the driving-field vector $\boldsymbol{\Omega}$, which consists of the static field $\boldsymbol{\Omega}_1$ in the x' axis, the rotating field $\boldsymbol{\Omega}_2(t)$ in the x'-y' plane with a frequency of $\Delta\omega_{12}=\omega_1-\omega_2$, and the static field $\Delta_1=\omega_0-\omega_1$ in the z' axis, as shown in Fig. 2.

In order to study the positions of the dominant resonances, we will first analyze the motion of the two-level system driven only by the saturating field. The saturating-laser field becomes time independent in the frame rotating at the frequency ω_1, and the atoms experience a static effective field in frequency units,

$$\Omega'=(\Delta_1^2+\Omega_1^2)^{1/2}\ . \tag{5}$$

The atomic-state vector $\boldsymbol{\rho}$ precesses around the driving-field vector $\boldsymbol{\Omega}$ at the characteristic Rabi frequency Ω'. The projection of this precession upon the x'-y' plane describes an ellipse, and can be decomposed into two components of different magnitudes rotating in opposite directions at the same frequency.

Going back to the laboratory frame the free precession contains therefore two frequencies, i.e., $\omega_1\pm(\Delta_1^2+\Omega_1^2)^{1/2}$. The second field will consequently change the population difference d_0 when its frequency ω_2 coincides with one of these two eigenfrequencies, that is, when the resonant condition

$$\omega_2=\omega_1\pm(\Delta_1^2+\Omega_1^2)^{1/2} \tag{6}$$

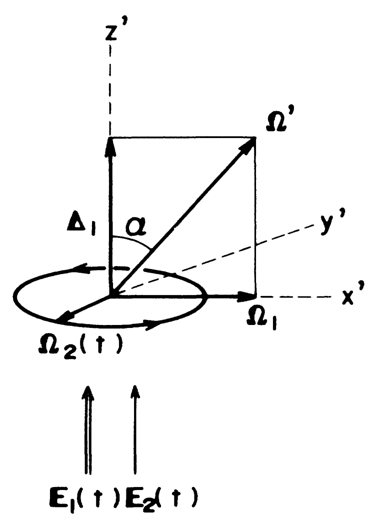

FIG. 2. Driving-field vectors in the frame rotating with the saturating-field frequency ω_1.

is satisfied.[4,13] When the saturating field is resonant ($\omega_0=\omega_1$), the two lines obtained are symmetric about the frequency $\omega_0=\omega_1$, their splitting being equal to $2\Omega_1$. When the saturating field is no longer resonant ($\omega_0\neq\omega_1$), the intensities of the two lines become unequal. It is easy to understand that the larger one is always closer to ω_0, as it must continuously evolve into the resonance line of the unperturbed atomic system when the detuning of the saturating field $\omega_0-\omega_1$ becomes very large. The two dominant peaks in each curve represented in Fig. 1 correspond to the resonances denoted by Eq. (6), and the small peaks that appear at $\omega_2-\omega_1=\pm\Omega/2$ are due to processes involving more than one second-laser photon (so-called Rabi subharmonics[6,9]).

A striking feature in Fig. 1 is that for $\omega_0-\omega_1$, the resonances at Rabi frequencies $\omega_2-\omega_1=\pm\Omega_1$ appear as an increase of the population difference d_0 rather than as a decrease, and when the pump field is slightly off resonant, i.e., for $\omega_0-\omega_1=5\gamma$, the population change exhibits a dispersion profile. It is expected in general that the external fields acting on the atomic system induce the stimulated transitions to reduce the population difference between the energy levels.[12] However, the results shown in Fig. 1 indicate that under some special conditions, the resonances appear as the increase of the population difference d_0 rather than as the decrease of it.

This peculiar effect has been discussed by Friedmann and Wilson-Gordon[14] as the competing processes of two-photon processes in which either a pump photon is absorbed and a probe photon is emitted or vice versa. These competing processes have no effect on absorption or stimulated emission of probe photons, but can only lead to stabilization of the ground state.

In what follows, we intend to explain the stabilization of the ground state as the transverse resonance which has been extensively studied in radio-frequency spectroscopy.

III. POPULATION RECOVERY BY THE SECOND-LASER FIELD

In order to investigate the detailed properties of the population recovery, the population difference d_0 is calculated for moderate and strong second-field intensities in the two-level atomic system driven by the strong, resonant saturating field ($\Omega_1=20\gamma$). This result is shown in Fig. 3, together with the absorption $\mathrm{Im}\rho_{ab}(\omega_2)$ and the dispersion $\mathrm{Re}\rho_{ab}(\omega_2)$ profiles for the second field. The zero levels for $\mathrm{Im}\rho_{ab}(\omega_2)$ and $\mathrm{Re}\rho_{ab}(\omega_2)$ are indicated by the dashed lines, and those for d_0 are abscissas in each drawing. We can see in Fig. 3 that for $\Omega_1\gg\Omega_2$ the maximum inhibition of the saturated transition, hence the maximum d_0, is obtained when $\omega_2-\omega_1\approx\pm\Omega_1$.

As has been pointed out by Friedmann and Wilson-Gordon,[14] the absorption profile of the second field $\mathrm{Im}\rho_{ab}(\omega_2)$ resembles two separate dispersion profiles, which is attributed to the competition between three- and four-photon scattering processes. The peaks of d_0 and of the dispersion curves $\mathrm{Re}\rho_{ab}(\omega_2)$ coincide with each other, and nearly correspond to the zeros of the absorption profiles $\mathrm{Im}\rho_{ab}(\omega_2)$ for $\Omega_1\gg\Omega_2$. It is worth noting that when the saturating field and the second field are the

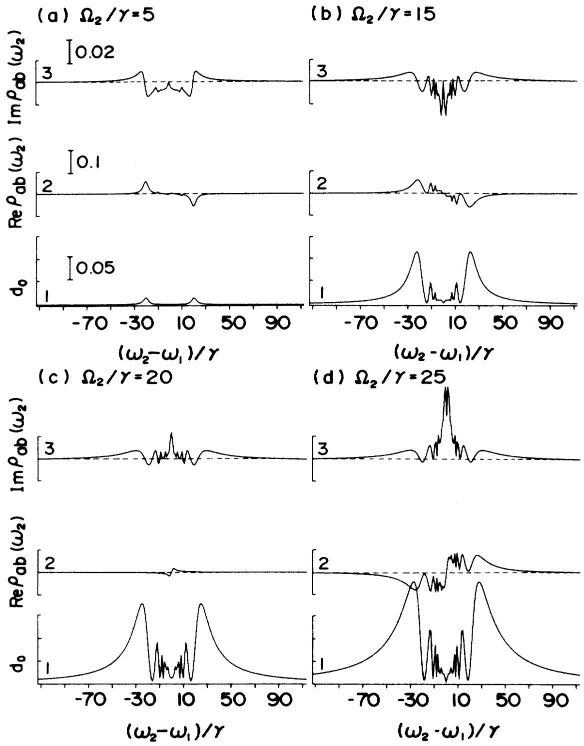

FIG. 3. Population difference d_0 calculated for various magnitudes of the second field in the two-level atomic system driven by the strong, resonant saturating fields ($\Omega_1/\gamma=20$). The corresponding dispersion $\mathrm{Re}\rho_{ab}(\omega_2)$ and absorption $\mathrm{Im}\rho_{ab}(\omega_2)$ profiles are also shown.

same amplitude, i.e., $\Omega_1=\Omega_2=20\gamma$, the dispersion curve has almost no structure, i.e., is dispersion free, except for a very small value of $(\omega_2-\omega_1)/\gamma$, as shown in Fig. 3(c). For $\Omega_1>\Omega_2$, the sign of the dispersion curve $\mathrm{Re}\rho_{ab}(\omega_2)$ changes from positive to negative when the value of $(\omega_2-\omega_1)/\gamma$ is varied from negative to positive, while for $\Omega_1<\Omega_2$ the polarity is interchanged. Consequently, the dispersion curve has no appreciable structure for $\Omega_1\approx\Omega_2$. The dispersion-free condition may be useful to obtain the phase matching in the interaction of two traveling waves.

In the central region ($|\omega_2-\omega_1|<\Omega_1$) there appear additional peaks whose amplitudes increase with increasing intensity of the second field. The peaks converge towards the center of the line ω_0, and their position is nearly described by the formula $|\omega_2-\omega_1|=\Omega_1/n$, $n=1,2,3,\ldots$, for the low and intermediate intensities of the second field. The peaks appearing within the Rabi sidebands have a width much less than the radiative linewidth, and should correspond to the "ultra-narrow" resonance observed by Bonch-Bruevich *et al.*[6] The peak value of the population difference d_0 is proportional to the square of the second-laser field amplitude, i.e., to the second-laser intensity, for the $\Omega_1\gg\Omega_2$, and then gradually saturates for $\Omega_1\lesssim\Omega_2$. The transition between the energy levels is almost completely saturated by the saturating field of $\Omega_1=20\gamma$ and $d_0\approx0.0025$. This nearly complete saturation is inhibited by the irradiation of the second-laser field, as shown in Fig. 3. The maximum values of the recovered population difference d_0 induced by the second field are 0.015, 0.06, 0.12, 0.18, and 0.23 for $\Omega_2/\gamma=5$, 10, 15, 20, and 25, respectively.

Mollow[4] has found that for the case of very intense saturating field exactly on resonance and lowest order in the second-field strength, the change of the population difference $\delta\bar{n}$ is given by the relation

$$\delta\bar{n}=|\Omega_2|^2\frac{\Delta_2^4-3(\Omega_1\Delta_2)^2-2(\Omega_1\gamma)^2}{(\Delta_2^2+\gamma^2)[(\Delta_2-\Omega_1)^2+\gamma^2][(\Delta_2+\Omega_1)^2+\gamma^2]}, \tag{7}$$

where $\Delta_2=\omega_0-\omega_2$. This expression predicts the peaks at $\Delta_2=\pm\Omega_1$, and the population change $\delta\bar{n}$ is proportional to the square of the second-field amplitude, i.e., the intensity of the second field. These properties coincide well with the results presented in Fig. 3 for the limited case of $\Omega_1\gg\Omega_2$. Mollow,[4] however, did not mention the concept of the inhibition of the stimulated transition by an extra-laser field, which is the principal discussion point of this paper.

IV. ANALOGY WITH MAGNETIC RESONANCE

In the following, we discuss the recovery of d_0 by the second field in connection with the magnetic resonance in the transverse optical pumping.[3] The fluorescent intensity of two-level atoms irradiated by a near-resonant amplitude-modulated light has been calculated in the Bloch model by Thomann.[15] He predicted the parametric resonances[16] that appear as the increase of the population difference d_0, and then pointed out the analogy with the magnetic resonance in a strong oscillating magnetic field making an arbitrary angle with the static field. Experimental evidence of the predicted parametric resonances has been obtained with an atomic beam of sodium. Although he has not mentioned the inhibition of the stimulated transition, the parametric resonances that appear as the increase of the population difference contain the same concept as the results predicted in this paper. The parametric resonance is one of the magnetic resonances due to transverse optical pumping.[16] The term "transverse" originates from the direction of the optical pumping with respect to that of the resultant magnetic field.

For the exact resonance condition of the saturating field ($\omega_1=\omega_0$), the resultant driving field consists of the static field $\mathbf{\Omega}_1$ and the rotating field $\mathbf{\Omega}_2(t)$ with an angular frequency $\omega_2-\omega_1$ in the x'-y' plane (see Fig. 2). This situation is exactly equivalent to that of the transverse-optical pumping considered by Tsukada[7] for different purpose, in which the atom is subjected to the static magnetic field $\mathbf{H}_0$ directed in y' axis, and the perturbing rotating field $\mathbf{H}_1(t)$ rotating in the plane containing $\mathbf{H}_0$, with the pumping beam perpendicular to $\mathbf{H}_0$ and $\mathbf{H}_1(t)$ (see Fig. 7 in Ref. 7). When the second field is not irradiated ($\Omega_2=0$), the two-level system is completely saturated by the strong saturating field, because the time-averaged z' component of the density matrix d_0 is given by $d_0=[1+(\Omega_1/\gamma)^2]^{-1}$, while for the presence of the second field ($\Omega_2\neq0$), the state vector $\boldsymbol{\rho}$ precesses around the resultant driving-field vector $[\mathbf{\Omega}=\mathbf{\Omega}_1+\mathbf{\Omega}(t)]$ in a complicated manner, and this motion creates the time-averaged z' component.

The structures of the resonance curves shown in Fig. 1 can be explained in terms of the competition of the longitudinal resonances and the transverse ones in the study of optical pumping.[17] The transverse resonance becomes dominant at $\omega_0=\omega_1$, so that the resonance structure appears as the increase of d_0. As the detuning of the saturating field is increased, the nature of the longitudinal resonance gradually increases and the resonances appear as the decrease of d_0. For the small detuning $(\omega_0-\omega_1=5\gamma)$, the curve shows the dispersion profile as the result of competition between the longitudinal and transverse resonances,[17,18] while for the large detuning, the resonance curve ultimately coincides with the resonance by the second field at $\omega_0=\omega_2$.

V. CONCLUSION

We showed that the large population recovery in the saturated two-level system is realized by the irradiation of the off-resonant extra-laser field. This effect was discussed in connection with the transverse-optical resonance which has been extensively studied in rf spectroscopy. It was also shown that the definite dispersion-free condition is realized when the intensities of the saturating field and the second field are the same.

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