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Resonance fluorescence and Autler-Townes spectra of a two-level atom driven by two fields of equal frequencies

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We study the effects of driving a two-level atom by two intense field modes that have equal frequencies but are otherwise distinguishable; the intensity of one mode is also assumed to be greater than that of the other. We calculate first the dressed states of the system, and then its resonance fluorescence and Autler-Townes absorption spectra. We find that the energy spectrum of the doubly dressed atom consists of a ladder of doublet continua. These continua manifest themselves in the fluorescence spectrum, where they produce continua at the positions of the Mollow sideband frequencies $\omega_L \pm 2\Omega$ of the strong field, and in the Autler-Townes absorption spectrum, which becomes a two-continuum doublet. [S1050-2947(97)01202-X]

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The radiative properties of two-level atoms driven by a bichromatic field have been the subject of intense theoretical and experimental investigation in recent years [1]. These investigations can be divided roughly into two categories: those in which the field components have (nearly) equal Rabi frequencies, and those in which the intensities differ significantly.

For nearly equal intensities, the fluorescence, absorption, and Autler-Townes spectra all consist of a series of features (equally) separated from each other by half the difference in frequency between the driving field components. The number of these features, their widths, and their relative intensities depend on the Rabi frequencies of the driving field. More recently, attention has turned to bichromatic excitation by one strong and one weak field component, and a different set of features have been both predicted [2] and observed, in Autler-Townes absorption [3] and in fluorescence [4]. In this case, multipeak features appear centered at ω_L and at the Mollow sideband frequencies $\omega_L \pm 2\Omega$ associated with the strong field component. The splittings and relative intensities within each multipeak feature depend in an intricate way on the detunings of the driving field components and on the ratio α of the Rabi frequency of the weak field to that of the strong. Furthermore, as α increases, additional features appear, centered at $\omega_L \pm n 2\Omega$, where *n* is an integer.

This novel behavior has led us to consider the case of bichromatic excitation by two field modes, one strong and one weaker, which have equal frequencies ω_L but are otherwise distinguishable. Our system consists of a two-level atom with excited state $|b\rangle$, ground state $|a\rangle$, and transition frequency ω_0 . The atom is driven by two field modes, whose frequencies ω_L are tuned close to ω_0 . The stronger (weaker) laser field has Rabi frequency 2Ω (2G). We use the doubly dressed atom model [5], first coupling the bare atom to the stronger field, next coupling the resulting "singly dressed" atom to the weaker field, and finally allowing the doubly dressed atom to decay spontaneously into the vacuum field. The model is valid in the limits $\omega_L \approx \omega_0 \ge 2\Omega > 2G > \Gamma$, where Γ is the rate of spontaneous emission to the vacuum modes.

The Hamiltonian H_{AL} of the atom+strong field mode has

eigenstates $|i,N\rangle$ (*i*=1,2), which satisfy the eigenvalue equation [6]

$$H_{AL}|i,N\rangle = \hbar \left[N\omega_L - \frac{\Delta}{2} - (-1)^i \Omega \right] |i,N\rangle, \qquad (1)$$

where

$$|1,N\rangle = \sin\theta |a,N\rangle + \cos\theta |b,N-1\rangle,$$

$$|2,N\rangle = \cos\theta |a,N\rangle - \sin\theta |b,N-1\rangle$$

(2)

are the (singly) dressed atom states with $|l,N\rangle(l=a,b)$ the state in which the atom is in state $|l\rangle$ and N photons are present in the field mode. Here, θ is given by

$$\cos^2\theta = \frac{1}{2} - \frac{\Delta}{4\Omega},\tag{3}$$

where $\Delta = \omega_L - \omega_0$, $2\Omega = (\Delta^2 + 4\Omega_1^2)^{1/2}$ is the detuned Rabi frequency, $2\Omega_1 = g_1 \sqrt{N}$ is the Rabi frequency at resonance, and g_1 is the atom-field coupling constant. The Hamiltonian of the second (weaker) field mode has the eigenvalue equation

$$H_W|n\rangle = n\hbar\,\omega_L|n\rangle,\tag{4}$$

and the noninteracting singly dressed-atom+weak field Hamiltonian $H_d = H_{AL} + H_W$ the eigenvalue equation

$$H_d|i,N-n;n\rangle = \hbar \left[N\omega_L - \frac{\Delta}{2} - (-1)^i \Omega \right] |i,N-n;n\rangle.$$
(5)

The states $|1,N-n;n\rangle(|2,N-n;n\rangle)$ $n=0,1,...,\infty$ form an infinite set of degenerate states with energy $\hbar[N\omega_L - \Delta/2 + \Omega](\hbar[N\omega_L - \Delta/2 - \Omega])$ [Fig. 1(a)].

When we include the interaction between the singly dressed atom and the weaker field

$$V = \hbar g_1 (a^+ S^- + S^+ a), \tag{6}$$

where $S^+(S^-)$ is the atomic raising (lowering) operator, the degeneracy is lifted. For $\Omega > G$ the coupling between the

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FIG. 1. Energy level spectra of the (a) singly dressed and (b) doubly dressed atom.

 $\{|1,N-n;n\rangle\}$ and $\{|2,N-n;n\rangle\}$ manifolds is negligible, and the interaction V has matrix elements $V_{nm}^{(i)}$ given by

$$V_{nm}^{(i)} = \langle i, N - n; n | V | i, N - m; m \rangle$$

= $-(-1)^{i} \hbar g(\sqrt{n+1} \delta_{n+1,m} + \sqrt{n} \delta_{n-1,m}),$ (7)

where $g = g_1 \sin\theta \cos\theta$. This matrix has the same form as that which represents the position operator in the basis provided by the energy eigenstates of the one-dimensional harmonic oscillator [7]. Its eigenvalues may be written as $-(-1)^i \lambda \hbar g$, $-\infty < \lambda < \infty$, and the total Hamiltonian of the system $\mathcal{H}_d + V$ satisfies the eigenvalue equation

$$(\mathcal{H}_d + V) |Ni\lambda\rangle = E_{N\lambda}^{(i)} |Ni\lambda\rangle, \qquad (8)$$

where

$$E_{N\lambda}^{(i)} = \hbar \left[N \omega_L - \frac{\Delta}{2} - (-1)^i (\Omega + \lambda g) \right], \tag{9}$$

$$|Ni\lambda\rangle = \sum_{n=0}^{\infty} \phi_n \left[-(-1)^i \frac{\lambda}{\sqrt{2}} \right] |i,N-n;n\rangle, \qquad (10)$$

and

$$\phi_n(x) = (\sqrt{2\pi} 2^n n!)^{-1/2} H_n(x) \exp\left(-\frac{1}{2}x^2\right).$$
(11)

In Eq. (11), $\phi_n(x)$ is the harmonic oscillator eigenfunction and $H_n(x)$ the Hermite polynomial of order *n*. The eigenvectors obey the orthonormality and completeness relations

$$\langle Ni\lambda | N'i'\lambda' \rangle = \delta_{NN'}\delta_{ii'}\delta(\lambda-\lambda')$$

and

$$\sum_{\mathbf{N},i} \int d\lambda |Ni\lambda\rangle \langle Ni\lambda| = 1.$$
 (12)

Electric dipole radiative transitions by the system are allowed between states in neighboring manifolds $\epsilon(N)$ and $\epsilon(N-1)$. Using the orthonormality and completeness relations, we easily verify that the nonvanishing matrix elements of the atomic raising operator S^+ are given by

$$\langle N1\lambda | S^+ | N-12\lambda' \rangle = (\cos^2\theta) \,\delta(\lambda + \lambda'), \quad (13a)$$

$$\langle N2\lambda|S^+|N-11\lambda'\rangle = -(\sin^2\theta)\,\delta(\lambda+\lambda'),$$
 (13b)

$$\langle Ni\lambda | S^+ | N-1i\lambda' \rangle = -(-1)^i (\sin\theta) (\cos\theta) \,\delta(\lambda - \lambda').$$
(13c)

The difference in sign within the arguments of the delta functions in the matrix elements corresponding to the triplet sidebands [Eqs. (13a), (13b)] and the central component [Eq. (13c)] is important, and originates from the parity of the eigenstates,

$$\phi_n(-x) = (-1)^n \phi_n(x).$$
(14)

The time evolution of the system is governed by the reduced atomic density operator ρ , which obeys the master equation

$$\frac{\partial \rho}{\partial t} = -\frac{i}{\hbar} \left[H, \rho \right] - \frac{1}{2} \Gamma (S^+ S^- \rho + \rho S^+ S^- - 2S^- \rho S^+), \tag{15}$$

where $H = H_d + V$. With this equation we can derive equations of motion for the reduced populations of the dressed states and the coherences between dressed states of neighboring manifolds. We consider first the situation in which the relative phase of the two driving field components is not kept constant throughout the experiment, and the driving field modes can therefore be represented by number states [6].

I. POPULATIONS

Following the method of Cohen-Tannoudji and Reynaud [8], we project the master equation (15) onto $|Ni\lambda\rangle$ on the right and $\langle Ni\lambda|$ on the left and sum over N; we thus obtain the following set of coupled equations for the populations $P_i(\lambda, t)$:

$$\dot{P}_{1}(\lambda,t) = -\Gamma(\cos^{4}\theta)P_{1}(\lambda,t) + \Gamma(\sin^{4}\theta)P_{2}(\lambda,t),$$

$$\dot{P}_{2}(\lambda,t) = -\Gamma(\sin^{4}\theta)P_{2}(\lambda,t) + \Gamma(\cos^{4}\theta)P_{1}(\lambda,t).$$
(16)

Here $P_i(\lambda,t) = \sum_N P_{iN}(\lambda,t)$, $P_{iN}(\lambda,t) = \langle Ni\lambda | \rho(t) | Ni\lambda \rangle$, and we assume that $P_{iN}(-\lambda,t) = P_{iN}(\lambda,t)$.

The steady-state solution of Eq. (16) is easily found to be

$$\frac{P_1(\lambda)}{P_2(\lambda)} = \frac{\sin^4\theta}{\cos^4\theta}.$$
(17)

We see that the population distribution between corresponding states of the two continua is the same as that between the dressed states of a two-level atom driven by a single laser field [6]. However, within each continuum the population distribution depends on λ : If there are *M* photons in laser mode 2, the populations $P_i(\lambda)$ are given by

$$P_{1}(\lambda) = \frac{\sin^{4}\theta}{\sin^{4}\theta + \cos^{4}\theta} \left| \phi_{M} \left(\frac{\lambda}{\sqrt{2}} \right) \right|^{2},$$
$$P_{2}(\lambda) = \frac{\cos^{4}\theta}{\sin^{4}\theta + \cos^{4}\theta} \left| \phi_{M} \left(\frac{\lambda}{\sqrt{2}} \right) \right|^{2}, \quad (18)$$

where $\phi_M(\lambda/\sqrt{2})$ is given by Eq. (10). The populations are a maximum for $\lambda/\sqrt{2}$ in the vicinity of the classical turning

points of the harmonic oscillator eigenfunction $\phi_M(\lambda/\sqrt{2})$, i.e., for $\lambda_M \sim \pm 2\sqrt{M+1/2}$, or for energies $|\lambda_M|\hbar g \approx 2\hbar g\sqrt{M} = 2(\sin\theta)(\cos\theta)\hbar G$. For $|\lambda| < |\lambda_M|$, the populations $P_i(\lambda)$ are smaller, but nonzero. For $|\lambda| > |\lambda_M|$, $P_i(\lambda)$ goes rapidly to zero.

To calculate the spectra, we require the time evolution of the average value of the raising part of the atomic dipole operator

$$\mu^{+} = \mu \sum_{i,j,N} \int \int d\lambda \ d\lambda' |Ni\lambda\rangle$$
$$\times \langle Ni\lambda | S^{+} | N - 1j\lambda' \rangle \langle N - 1j\lambda' |$$
$$= \mu \sum_{i,j,N} \int \int d\lambda \ d\lambda' \mu_{ij}(\lambda,\lambda') \rho_{ijN}^{(+)}(\lambda,\lambda'), \quad (19)$$

where $\mu_{ij}(\lambda, \lambda') = \langle Ni\lambda | S^+ | N - 1j\lambda' \rangle$ and

$$\rho_{ijN}^{(+)} = |Ni\lambda\rangle\langle N - 1j\lambda'| \tag{20}$$

is an off-diagonal element of the density operator. Since the continua of the dressed states do not overlap, we can apply the secular approximation [6,8], which allows us to consider transitions between the two manifolds as appearing at three significantly different frequencies. We consider first the average dipole moment at $\omega_L + 2\Omega$, by projecting the master equation (15) onto $|N-12\lambda'\rangle$ on the right and $\langle N1\lambda|$ on the left and then summing over N. This results in the equation of motion

$$\dot{\rho}_{12}(\lambda,\lambda') = -\left\{ \Gamma\left((\sin^2\theta)(\cos^2\theta) + \frac{1}{2} \right) + i[\omega_L + 2\Omega + (\lambda - \lambda')g] \right\} \rho_{12}(\lambda,\lambda'),$$
(21)

where $\rho_{12}(\lambda,\lambda') = \sum_N \rho_{12N}^{(+)}(\lambda,\lambda')$. Similarly, the dipole moment at $\omega_L - 2\Omega$ evolves according to the equation

$$\dot{\rho}_{21}(\lambda,\lambda') = -\left\{ \Gamma\left((\sin^2\theta)(\cos^2\theta) + \frac{1}{2} \right) + i[\omega_L - 2\Omega + (\lambda - \lambda')g] \right\} \rho_{21}(\lambda,\lambda').$$
(22)

For the central component at ω_L , we project the master equation (15) onto $|N-1i\lambda'\rangle$ on the right and $\langle Ni\lambda|$ on the left (i=1,2), and sum over N. This results in the following two coupled equations of motion:

$$\dot{\rho}_{11}(\lambda,\lambda') = -\{\Gamma(\cos^4\theta) + i[\omega_L + (\lambda - \lambda')g]\}\rho_{11}(\lambda,\lambda') + \Gamma(\sin^4\theta)\rho_{22}(\lambda,\lambda'),$$
(23)

$$\dot{\rho}_{22}(\lambda,\lambda') = -\{\Gamma(\sin^4\theta) + i[\omega_L + (\lambda - \lambda')g]\}\rho_{22}(\lambda,\lambda') + \Gamma(\cos^4\theta)\rho_{11}(\lambda,\lambda').$$

II. FLUORESCENCE SPECTRUM

The fluorescence spectrum is given by the real part of the Fourier transform of the two-time correlation function of the dipole-moment operator $\langle \mu^+(t+\tau)\mu^-(t)\rangle$, $\tau>0$. From the quantum regression theorem [9], it is well known that for $\tau>0$ the two-time average $\langle \mu^+(t+\tau)\mu^-(t)\rangle$ satisfies the same equation of motion as the one-time average $\langle \mu^+(\tau)\rangle$. The equation of motion for $\langle \mu^+(\tau)\rangle$ is obtained from Eq. (19) in the form

$$\langle \dot{\mu}^{+}(\tau) \rangle = \int \int d\lambda \ d\lambda' \{ (\sin\theta)(\cos\theta) [\dot{\rho}_{11}(\lambda,\lambda') - \dot{\rho}_{22}(\lambda,\lambda')] \delta(\lambda - \lambda') + (\cos^2\theta) \dot{\rho}_{12}(\lambda,\lambda') \delta(\lambda + \lambda')$$

- $(\sin^2\theta) \dot{\rho}_{21}(\lambda,\lambda') \delta(\lambda + \lambda') \},$ (24)

where $\dot{\rho}_{ij}(\lambda,\lambda')$ are given in Eqs. (21)–(23). The spectrum is then obtained as the real part of the Fourier transform of the two-time correlation function $\langle \mu^+(t+\tau)\mu^-(t)\rangle$, and is given by

$$S(\omega) = \frac{\Gamma}{\pi} \left\{ (\sin^2 \theta) (\cos^2 \theta) \left[\pi \delta(\omega - \omega_L) \frac{(\sin^4 \theta - \cos^4 \theta)^2}{(\sin^4 \theta + \cos^4 \theta)^2} + \left(\frac{2(\sin^2 \theta) (\cos^2 \theta)}{\sin^4 \theta + \cos^4 \theta} \right)^2 \frac{\Gamma_p}{(\omega - \omega_L)^2 + \Gamma_p^2} \right] + \frac{(\sin^4 \theta) (\cos^4 \theta)}{(\sin^4 \theta + \cos^4 \theta)} \int_{-\infty}^{+\infty} d\lambda |\phi_M(\lambda/\sqrt{2})|^2 \left(\frac{\Gamma_c}{(\omega - \omega_L - 2\Omega - 2\lambda g)^2 + \Gamma_c^2} + \frac{\Gamma_c}{(\omega - \omega_L + 2\Omega - 2\lambda g)^2 + \Gamma_c^2} \right) \right\}, \quad (25)$$

where $\Gamma_c = \Gamma[(\sin^2\theta)(\cos^2\theta) + 1/2]$ and $\Gamma_p = \Gamma(\sin^4\theta + \cos^4\theta)$.

We find that the presence of the second weaker field of the same frequency as the first leads to a modification of the Rabi sidebands, while the central component remains unchanged. In Fig. 2, we plot $S(\omega)$ for $\Delta=0$, $2\Omega=40\Gamma$, and different G. The spectrum exhibits a central component, identical to that of the Mollow triplet, together with broadened sideband continua centered at $\omega_L \pm 2\Omega$. Each sidebandconsists of a convolution of Lorentzian functions, centered at $\omega_L \pm 2\Omega + 2\lambda g$ and having width Γ_c , multiplied by a weight factor $|\phi_M(\lambda/\sqrt{2})|^2$, reflecting the steady-state populations of the eigenstates of energy $\pm \hbar \Omega + \lambda \hbar g$. Because $|\phi_M(\lambda/\sqrt{2})|^2$ reaches a maximum near the classical turning points $|\lambda_M| \sim 2\sqrt{M}$ and then goes rapidly to zero, the sidebands display peaks near $\omega \sim \omega_L \pm 2\Omega \pm 2G$, with continua between the



FIG. 2. The fluorescence spectrum $S(\omega)$ for $\omega_L = \omega_0$, $2\Omega = 40\Gamma$, and (a) 2G = 7.1, (b) 2G = 14.2.

peaks, and then go rapidly to zero.

We point out that the absence of change in the central component of the spectrum is ultimately traceable (mathematically) to the minus sign in the argument of the delta function in the transition elements of Eq. (13c), while the broadening of the sidebands arises from the plus sign in the delta functions in Eqs. (13a) and (13b).

III. AUTLER-TOWNES ABSORPTION SPECTRUM

If the system is probed by tuning a low-intensity laser in the vicinity of the transition frequency between one of the resonantly coupled atomic levels $(|a\rangle, \text{ say})$ and a third atomic level $(|c\rangle)$, the Autler-Townes absorption spectrum is observed. For a monochromatic intense driving field, this spectrum consists of a doublet, separated by the Rabi frequency 2Ω of the field [6]. The weight of each component is proportional to the product of the steady-state population of the level $|i,N\rangle$ from which the absorption originates and the transition rate from $|i,N\rangle$ to $|c,N\rangle$, proportional to $|\langle a,N|i,N\rangle|^2$.

Analogously, in our bichromatically driven system, the Autler-Townes spectrum is calculated to be

$$A(\omega) = \frac{\Gamma'}{\pi} \frac{1}{\sin^4 \theta + \cos^4 \theta} \int_{-\infty}^{\infty} d\lambda |\phi_M(\lambda \sqrt{2})|^2 \\ \times \left[\frac{\sin^4 \theta (\Gamma' + \Gamma_1)/2}{(\omega - \omega_{ca} - \Delta/2 + \Omega - \lambda g)^2 + \frac{1}{4} (\Gamma' + \Gamma_1)^2} \right. \\ \left. + \frac{\cos^4 \theta (\Gamma' + \Gamma_2)/2}{(\omega - \omega_{ca} - \Delta/2 - \Omega - \lambda g)^2 + \frac{1}{4} (\Gamma' + \Gamma_2)^2} \right]$$
(26)



FIG. 3. The Autler-Townes absorption spectrum $A(\omega)$ for $\omega_L = \omega_0$, $2\Omega = 40\Gamma$, and (a) 2G = 7.1, (b) 2G = 14.2.

where Γ' is the natural width of level $|c\rangle$, $\Gamma_1 = \Gamma \cos^2 \theta$ and $\Gamma_2 = \Gamma \sin^2 \theta$. Each doublet feature consists of a convolution of Lorentzian lines, with each line weighted by the steady-state population $P_i(\lambda)$ of the level from which the absorption originates and the transition rate to $|c\rangle$. In Fig. 3 we plot the Autler-Townes spectra for $\Delta = 0$, $2\Omega = 40\Gamma$, and different *G*. We see that in this case each doublet component is broadened into a continuum centered at $\omega_{ca} + \Delta/2 \pm \Omega$, consisting of a convolution of appropriately weighted Lorentzian functions. These continua display maxima near $\omega_{ca} + \Delta/2 \pm \Omega \pm \Omega$, and then rapidly go to zero.

In summary, we have investigated analytically the fluorescence and Autler-Townes spectra of a two-level atom driven by two fields of equal frequencies. The spectra differ qualitatively from both the monochromatic driving field case and the bichromatic case with two fields of unequal frequencies [1-4]. In the fluorescence spectrum, the presence of a weaker field of the same frequency leads to a broadening of the Rabi sidebands of the strong field into continua of width $\sim 4G$, whereas the central component remains unchanged. In the Autler-Townes spectrum, the doublet components are broadened into continua of width $\sim 2G$. We have interpreted the spectral features in terms of the dressed states of the system and transitions among them: These dressed states are grouped within two continua separated by the Rabi frequency of the strong field; the bandwidth of each continuum is determined by the Rabi frequency of the weak field.

An experiment was performed recently [3] studying the Autler-Townes spectrum of a probe beam monitoring a twolevel transition of a three-level vee system driven by two laser fields of the same frequency, but significantly different intensities and uncoordinated relative phase. The observed spectrum was composed of two continuum structures sepa-

IV. FIXED RELATIVE PHASE OF THE DRIVING FIELD COMPONENTS

It is interesting to regard this problem from the point of view of fluctuating driving fields. In Ref. [3], the experiments were performed using two driving fields, each intensity and phase stabilized so that for each mode the correlation times for both slow and fast fluctuations, τ_d and τ_c , respectively [10], were very large: $G \gg \tau_d^{-1}$, τ_c^{-1} . However, the phases of the modes were not interlocked, and signal averaging was employed, so that their relative phase was random. In this situation, it is sufficient in practice to represent the driving field (two uncorrelated phase-diffusion modes, each with zero bandwidth) by number (Fock) states, rather than requiring a coherent (Glauber) state representation: They produce the same results [11]. Very recently, however, Autler-Townes absorption spectra were observed with the relative phases of the field modes kept coordinated and no signal averaging employed. Furthermore, the frequency of the weaker field was $\omega_2 = \omega_L + \delta$, where, $\delta \ll \omega_L$, Γ [12]. To describe this situation theoretically, the field modes must be represented by coherent states. The results of this calculation can be described simply, however, and are (also) in agreement with the experimental observations (which will

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be reported in detail elsewhere): The average electric field of mode 2 can be written

$$\vec{E} = \vec{E}_2 \cos[(\omega_L + \delta)t]$$
$$= (\vec{E}_2 \cos \delta t) \cos \omega_L t + (\vec{E}_2 \sin \delta t) \sin \omega_L t.$$
(27)

Because $\delta \ll \omega_L$, Γ (of order Hertz, experimentally), each atom follows the $\cos \delta t$ (and $\sin \delta t$) time behavior adiabatically. For optical-frequency driving fields, the wavelength λ is much smaller than the dimensions of the sample, and the spectra must still be averaged over the different relative phases experienced by the different atoms: Again, the spectra will involve continua, as with random relative phases. For radio-frequency driving fields however, the atoms experience the same relative phase. For $\delta = 0$, a normal Autler-Townes doublet is observed, with Rabi frequency corresponding to the (vector) sum of the two electric fields. For small but nonzero δ , the doublet components are observed to move about the central frequencies $\omega_{ca} + \Delta/2 \pm \Omega$, with amplitude *G* [12].

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