ac Stark splitting in double optical resonance and resonance fluorescence by a nonmonochromatic chaotic field

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A detailed theoretical analysis of ac Stark splitting in double optical resonance and resonance fluorescence by a nonmonochromatic complex chaotic field is presented. It is shown that a one-photon transition undergoes Stark splitting, which is strongly affected by the intensity fluctuations of the chaotic light. On resonance, the lines in the doublet structure of double optical resonance and the sidebands of the triplet structure in resonance fluorescence are broadened by the intensity fluctuations and tend to copy the Rayleigh distribution of the field amplitude. Off resonance, the asymmetry of the doublet in double optical resonance is reversed and a sideband asymmetry appears in resonance fluorescence for finite bandwidth of the incident light. While on resonance the splitting is always reduced, off resonance the intensity fluctuations may lead to an enhancement of the splitting in comparison to phase-diffusing light.

I. INTRODUCTION

Until recently, the theory of the resonant interaction of intense laser radiation with atoms had been based mainly on the assumption of monochromatic and coherent excitation. Although neglecting the bandwidth and intensity fluctuations of the laser seems to be justified for the interpretation of recent experiments employing well-stabilized cw lasers,[1-3] light from pulsed multimode lasers exhibits large intensity fluctuations and often has a bandwidth at least comparable to the relaxation constants of the atom.[4] The interpretation of experiments performed with such lasers requires a generalized theory which can account for the fluctuations of the exciting light. As has already been apparent in recent experiments and as is shown in this paper, the fluctuations of the field and the associated bandwidth introduce important new effects.

For a nonresonant N-photon transition to a broad final state, which is described by a rate in Nth order perturbation theory, the effect of field fluctuations is well understood as it enters through the Nth order correlation function of the incident light.[5] The calculation of transition probabilities and line shapes of saturated resonant processes, however, requires the summation of an infinite sequence of resonant terms of the perturbation series and contains therefore information about all higher-order correlation functions of the field.[6] This of course stems from the fact that saturation is a highly nonlinear process. Under such conditions, general solutions for electromagnetic fields of arbitrary stochastic properties are no longer available. The problem of atom-field interaction must then be solved separately for each model of the stochastic field. Two particular models, the phase-diffusion (PD) model[7,8] corresponding to a well-stabilized cw laser with a diffusing phase and the chaotic field (CF)[7] describing thermal light or light from a multimode laser with a large number of independent modes,[9] have attracted particular interest in this context. The effect of a finite laser bandwidth on the basis of the PD model is by now well understood.[10-15] On the other hand, the understanding of these effects in CF, owing to serious mathematical difficulties, has been either purely qualitative[13] or based on such methods as the decorrelation approximation (DA),[11,16-18] the validity of which for the CF in the saturation regime is at best questionable. It is only recently that solutions for the stationary density-matrix elements of a two-level atom (TLA) in a CF field have been given.[19,20] Double-resonance and resonance fluorescence of a TLA in a CF have, however, remained unsolved problems in this area.

As is well known, an atomic transition, under the influence of a coherent resonant intense field, undergoes Stark splitting when the Rabi frequency becomes larger than the natural decay widths of the atom.[21] This splitting can be observed either in double resonance[3,22,23] or in fluorescence.[1,2,24-26] In double resonance, the TLA is probed by a second weak laser, inducing transitions to a third unperturbed level.[4] The splitting of the resonance line into a doublet, with peaks separated by the Rabi frequency of the TLA, is observed as a function of the probe frequency. In resonance fluorescence, the spectrum of the TLA exhibits a triplet structure.[1,2] The central line coincides with the laser frequency and is accompanied by two sidebands which, for on-resonance excitation, are separated by the Rabi frequency. Since the Rabi frequency is proportional to the electric field amplitude of the incident laser light, the large intensity fluctuations of the CF, which are of the order of the intensity itself, will tend to wash out the splitting. On the basis of such qualitative arguments, it has been predicted that the triplet spectrum would disappear for CF's.[13] On the other hand, calculations within somewhat simplified models predict ac Stark splitting in double resonance.[27,28] The splitting of spectral lines in resonance fluorescence and in double optical resonance are, however, closely related. In view of these discrepancies in the predictions for the spectrum of double resonance and resonance fluorescence in CF's, there is a need for accurate calculations capable of answering questions regarding the existence of ac Stark splitting for a TLA, the magnitude of the splitting, and the widths and heights of the spectral lines of the spectrum.

In this paper I calculate the spectrum of double resonance and resonance fluorescence in an intense nonmonochromatic CF within a formalism that has been developed in a series of papers.[14,19,29,30] Starting from a stochastic model for the CF, introduced in Sec. II, I derive equations for the averaged atomic populations in double resonance within the weak-probe approximation in Sec. III. Within a similar approach Sec. IV gives a treatment of the spectrum of resonance fluorescence.

II. MODEL FOR THE CHAOTIC FIELD

A multimode laser with a large number of independent modes is accurately described by a nonmonochromatic ideal CF.[9] This model has also been employed by other authors in the study of different processes.[17,18] Typically, the Nth-order correlation function of the electric CF amplitude $\epsilon(t)$ satisfies[7]

$$\langle \epsilon^*(t_1)\cdots\epsilon^*(t_n)\epsilon(t_{n+1})\cdots\epsilon(t_{2n})\rangle = \sum_P \prod_{j=1}^{n} \langle \epsilon^*(t_j)\epsilon(t_{P(j+n)})\rangle , \quad (1)$$

where P stands for permutation. Assuming the spectrum to be Lorentzian, the first-order correlation function is given by $\langle\epsilon^*(t)\epsilon(t')\rangle = \langle|\epsilon|^2\rangle e^{-b|t-t'|}$, with b being the bandwidth of the spectrum.

A stochastic model for $\epsilon(t)$ having the above properties of a CF can be described in terms of the Langevin equations[30]

$$\dot{\epsilon}(t) = -b\epsilon(t) + F_\epsilon(t) , \quad \dot{\epsilon}^*(t) = -b\epsilon^*(t) + F_{\epsilon^*}(t) , \quad (2)$$

with Gaussian random forces[31]

$$\langle F_\epsilon(t)F_{\epsilon^*}(t')\rangle = 2b\,\langle|\epsilon|^2\rangle\delta(t-t')$$

and

$$\langle F_\epsilon(t)F_\epsilon(t')\rangle = \langle F_{\epsilon^*}(t)F_{\epsilon^*}(t')\rangle = 0 .$$

Thus we assume that $\epsilon(t)$ obeys a normal Markov process.[31] The Fokker-Planck operator $L(\epsilon,\epsilon^*)$ in the corresponding master equation

$$[(\partial/\partial t) + L(\epsilon,\epsilon^*)]P(\epsilon,\epsilon^*,t) = 0$$

for the probability distribution $P(\epsilon,\epsilon^*,t)$ is given by[30,31]

$$L(\epsilon,\epsilon^*) = -b\left(\frac{\partial}{\partial\epsilon}\,\epsilon + \frac{\partial}{\partial\epsilon^*}\,\epsilon^* + 2\langle|\epsilon|^2\rangle\frac{\partial^2}{\partial\epsilon\partial\epsilon^*}\right). \quad (3)$$

The stationary solution $LP_s(\epsilon,\epsilon^*)=0$ of the master equation is the Glauber P-distribution function for the CF[7]

$$P_s(\epsilon,\epsilon^*) = (1/\pi\langle|\epsilon|^2\rangle)e^{-|\epsilon|^2/\langle|\epsilon|^2\rangle} . \quad (4)$$

III. DOUBLE RESONANCE

We consider an atom with ground state $|0\rangle$ and two excited states $|1\rangle$ and $|2\rangle$, with respective energies $\hbar\omega_0 < \hbar\omega_1 < \hbar\omega_2$.[15] The transitions $|0\rangle - |1\rangle$ and $|1\rangle - |2\rangle$ are dipole allowed, while $|0\rangle - |2\rangle$ is forbidden by parity-selection rules. The excited states $|1\rangle$ and $|2\rangle$ have natural decay widths K_1 and K_2. We assume that the first transition $|0\rangle - |1\rangle$ is strongly driven by the CF of Sec. II. In double resonance the ac Stark splitting of this transition is detected by observing the population induced in level $|2\rangle$ by a weak-probe laser as a function of the probe frequency.[4] This population can be measured either by monitoring its fluorescence to some unperturbed level or by observing the total ionization from this uppermost level.[4] In order not to perturb the Stark splitting of the strongly driven TLA $|0\rangle$, $|1\rangle$, we neglect the influence of ionization and adopt the weak-probe approximation, i.e., we assume that the probe laser does not affect the strongly driven TLA $|0\rangle$, $|1\rangle$ ($\rho_{11} + \rho_{00} = 1; \rho_{22} \approx 0$). If the ionization or the probe is strong, a much more complicated structure appears which is of no interest in this paper. In this way we obtain the following equations for the slowly varying density-matrix elements in the rotating-wave approximation[15,22,23]:

$$\left(\frac{d}{dt} + K_2\right)\rho_{22}(t) = i\tfrac{1}{2}\Omega'\rho_{12}(t) + \text{c.c.} ,$$

$$\left(\frac{d}{dt} + i\Delta_2 + \tfrac{1}{2}K_{12}\right)\rho_{12}(t) = -i\tfrac{1}{2}\Omega'\rho_{11}(t) + i\mu_{01}\epsilon(t)\rho_{02}(t) , \quad (5)$$

$$\left(\frac{d}{dt} + i\Delta_1 + i\Delta_2 + \tfrac{1}{2}K_{02}\right)\rho_{02}(t) = -i\tfrac{1}{2}\Omega'\rho_{01}(t) + i[\mu_{01}\epsilon(t)]^*\rho_{12}(t) .$$

ρ_{00}, ρ_{12}, and ρ_{11} fulfill the equations of the TLA coupled to the strong field but not to the probe:

$$\left(\frac{d}{dt}+K_1\right)\rho_{11}(t)=i\mu_{01}\epsilon(t)\rho_{01}(t)+\text{c.c.}\,,$$
$$\left(\frac{d}{dt}+i\Delta_1+\tfrac{1}{2}K_{01}\right)\rho_{01}(t) = i[\mu_{01}\epsilon(t)]^*[\rho_{11}(t)-\rho_{00}(t)]\,, \quad (6)$$
$$\rho_{11}(t)+\rho_{00}(t)=1\,,$$

with $K_{ij}=K_i+K_j$ and μ_{01} being the dipole matrix element of the transition $|0\rangle$-$|1\rangle$ and Ω' the Rabi frequency of the probe. $\Delta_1=\omega-\omega_{10}$ and $\Delta_2=\omega' - \omega_{21}$ correspond to the detuning in the transitions $|0\rangle$-$|1\rangle$ and $|1\rangle$-$|2\rangle$, respectively. A Lorentzian bandwidth b' of the weak-probe laser is easily included in Eq. (5) by the substitutions $\frac{1}{2}K_{12}+b'$ and $\frac{1}{2}K_{02}+b'$.[14,15]

Since the electric field amplitude $\epsilon(t)$ is a stochastic function of time, the density-matrix equations (5) and (6) are stochastic differential equations[32] which must be solved for the averaged population $\langle\rho_{22}(t)\rangle_t$ of level $|2\rangle$. $\langle\ \rangle_t$ denotes averaging over the fluctuations in the incident field. The subscript indicates the explicit time dependence of this average. Equations (5) show that the population of state $|2\rangle\,\rho_{22}(t)$, as induced by the weak probe, depends on the off-diagonal density-matrix element $\rho_{12}(t)$. Because of the fluctuating amplitude, $\rho_{12}(t)$ and $\rho_{02}(t)$ become stochastic functions of time. The stochastic behavior is induced directly by $\epsilon(t)$ as well as by the fluctuations in the population $\rho_{11}(t)$ and the off-diagonal density-matrix element of the TLA $\rho_{01}(t)$. Both of these fluctuations, of course, have their origin in the stochastic driving field.

In the limit of a field with zero bandwidth, the amplitude $\epsilon(t)$ is no longer a stochastic function of time, but becomes a time-independent random variable distributed according to the stationary distribution function $P_s(\epsilon,\epsilon^*)$.[32] The averaged density-matrix elements $\langle\rho_{ij}(t)\rangle_t$ may therefore be found by solving Eqs. (5) and (6) with a constant field amplitude $\epsilon(t)=\epsilon$ for $\rho_{ij}^{b=0}(\epsilon,\epsilon^*,t)$, which is the solution for a monochromatic coherent driving field, and averaging this solution over the probability function P_s[30]:

$$\langle\rho_{ij}(t)\rangle_t^{b=0} = \int d^2\epsilon\, P_s(\epsilon,\epsilon^*)\rho_{ij}^{b=0}(\epsilon,\epsilon^*,t)\,. \quad (7)$$

With the help of the stationary solution for the population of level $|2\rangle$ in coherent fields as has been given in Ref. 22 [see also Eq. (14) below], we find

$$\langle\rho_{22}(t)\rangle=\frac{1}{2}\frac{\Omega'^2}{K_2}\frac{T+R^*}{\Omega^2}\times\frac{1}{a-b}\left[ae^aE_1(a)-be^bE_1(b)\right]+\text{c.c.}\,, \quad (8)$$

with $a=4ST/\Omega^2$, $b=2|R|^2/\Omega^2$, and $\Omega=2\mu_{10}\langle|\epsilon|^2\rangle^{1/2}$. R, S, and T are defined by $R=i\Delta_1+\frac{1}{2}K_{01}$, $S=i\Delta_2+\frac{1}{2}K_{12}$, and $T=i(\Delta_1+\Delta_2)+\frac{1}{2}K_{02}$. E_1 denotes the exponential integral.[33]

The solution of the density-matrix equation for finite bandwidth fields is one of the central difficulties in the treatment of the interaction of atoms with stochastically fluctuating fields. One commonly employed approximation scheme in solving these equations has been the decorrelation approximation (DA) which decorrelates atom-field averages of the form[11,16-18]

$$\langle\epsilon(t)\epsilon^*(t')\rho_{ii}(t')\rangle\approx\langle\epsilon(t)\epsilon^*(t')\rangle\langle\rho_{ii}(t')\rangle\,.$$

This decorrelation can be justified only in the limit in which the fluctuations of the electric field are much faster than those of the populations, i.e., the bandwidth of the field $\epsilon(t)$ must be larger than the Rabi frequency $\Omega=2\mu_{10}\langle|\epsilon(t)|^2\rangle^{1/2}$ (Ref. 32). The DA is therefore only valid in the weak-field limit and becomes questionable when the driving field becomes sufficiently intense to saturate the atomic transition.

We have recently outlined a rigorous method of solving the density-matrix equations for a stochastic Markovian driving field. Details of this method are given in Ref. 30. A short outline of the application of this formalism to Eq. (5) may be found in the Appendix. The idea of the method is to convert the stochastic density-matrix equation to an infinite system of differential equations of certain atom-field averages depending only on one time argument.

Applying this formalism to the density-matrix equation (5) and using Eq. (A8) of the Appendix, we find that for the CF of Sec. 2 the atom-field averages

$$\rho_{ij}^{\alpha n}(t)=\left(\frac{n!}{(n+|\alpha|)!}\right)^{1/2}\langle|\epsilon(t)|^{|\alpha|}[\epsilon(t)/\epsilon^*(t)]^{\alpha/2}L_n^{|\alpha|}(|\epsilon(t)|^2/\langle|\epsilon|^2\rangle)\rho_{ij}(t)\rangle/\langle|\epsilon|^2\rangle^{|\alpha|/2}\,,$$
$$\alpha=0,\pm1,\ldots,\quad n=0,1,2,\ldots, \quad (9)$$

with L_n^α being Laguerre polynomials[33] obey the infinite system of differential equations

$$\left(\frac{d}{dt}+K_2+2nb\right)\rho_{22}^{0n}(t)=i\tfrac{1}{2}\Omega'\rho_{12}^{0n}(t)+\text{c.c.}\,,\quad n=0,1,2,\ldots,$$

$$\left(\frac{d}{dt}+S^n\right)\rho_{12}^{0n}(t)=i\tfrac{1}{2}\Omega\left[\sqrt{n+1}\,\rho_{02}^{1n}(t)-\sqrt{n}\,\rho_{02}^{1n-1}(t)\right]-i\tfrac{1}{2}\Omega'\rho_{11}^{0n}(t)\,,$$

$$\left(\frac{d}{dt}+T^n\right)\rho_{02}^{1n}(t)=i\tfrac{1}{2}\Omega\sqrt{n+1}\left[\rho_{12}^{0n}(t)-\rho_{12}^{0n+1}(t)\right]-i\tfrac{1}{2}\Omega'\rho_{01}^{1n}(t)\,, \tag{10}$$

$$\left(\frac{d}{dt}+K_1+2nb\right)\rho_{11}^{0n}(t)=-i\tfrac{1}{2}\Omega\left[\sqrt{n+1}\,\rho_{01}^{1n}(t)-\sqrt{n}\,\rho_{01}^{1n-1}(t)\right],$$

$$\left(\frac{d}{dt}+R^n\right)\rho_{01}^{1n}(t)=i\tfrac{1}{2}\Omega\sqrt{n+1}\left[\rho_{11}^{0n}(t)-\rho_{00}^{0n}(t)-\rho_{11}^{0n+1}(t)+\rho_{00}^{0n+1}(t)\right],$$

$$\rho_{11}^{0n}(t)+\rho_{00}^{0n}(t)=\delta_{n,0}\,,$$

with $\Omega=2\mu_{01}\sqrt{\langle|\epsilon|^2\rangle^{1/2}}$ being the Rabi frequency of the transition $|0\rangle-|1\rangle$,

$$R^n=i\Delta_1+\tfrac{1}{2}K_{01}+(2n+1)b\,,\quad S^n=i\Delta_2+\tfrac{1}{2}K_{12}+2nb\,,$$

and $T^n=i\Delta_1+i\Delta_2+\frac{1}{2}K_{02}+(2n+1)b$. Note that the averaged populations are simply given by $\langle\rho_{ii}(t)\rangle_t=\rho_{ii}^{00}(t)$.

Confining ourselves to the stationary limit and neglecting time derivatives, Eq. (10) gives the following expression for the population of level $|2\rangle$:

$$\langle\rho_{22}(t)\rangle=i\frac{1}{2}\frac{\Omega'}{K_2}\rho_{12}^{00}+\text{c.c.}\,, \tag{11}$$

where ρ_{12}^{0n} fulfills the inhomogeneous three-term recursion formula

$$\left(S^n+\frac{1}{4}\frac{(n+1)\Omega^2}{T^n}+\frac{1}{4}\frac{n\Omega^2}{T^{n-1}}\right)\rho_{12}^{0n}-\frac{1}{4}\frac{(n+1)\Omega^2}{T^n}\rho_{12}^{0n+1}-\frac{1}{4}\frac{n\Omega^2}{T^{n-1}}\rho_{12}^{0n-1}$$
$$=-i\tfrac{1}{2}\Omega'\rho_{11}^{0n}+\tfrac{1}{4}\Omega'\Omega\left(\frac{\sqrt{n+1}}{T^n}\rho_{01}^{1n}-\frac{\sqrt{n}}{T^{n-1}}\rho_{01}^{1n-1}\right),\quad n=0,1,2,\ldots. \tag{12}$$

The coupling of the average ρ_{12}^{00} to all the other higher-order atom-field averages ρ_{12}^{0n} indicates explicitly the influence of all higher-order correlation functions of the incident field. The inhomogeneous terms on the right-hand side of Eq. (12) arise from the population and population fluctuations in the system $|0\rangle$, $|1\rangle$. Note that the differences $W^n=\rho_{11}^{0n}-\rho_{00}^{0n}$ [from which ρ_{11}^{0n} and ρ_{01}^{1n} may be found with the help of Eq. (10)] fulfill a similar recursion relation

$$(K_1+2nb+A_n+A_{n-1})W^n-A_nW^{n+1}-A_{n-1}W^{n-1}=-K_1\delta_{n0}\,, \tag{13}$$

with

$$A_n=(n+1)\Omega^2\frac{\frac{1}{2}K_{01}+(2n+1)b}{\Delta_1^2+\left[\frac{1}{2}K_{01}+(2n+1)b\right]^2}\,.$$

As we discussed in Ref. 19, the solution of Eq. (13) can be found in terms of continued fractions so we can assume that the right-hand side of Eq. (12) is known. On resonance and for high intensities the terms in Eq. (12), corresponding to the population fluctuations, can be neglected. Such an approach has been used in Ref. 20. Our method includes these terms which will be seen to have a significant effect on the off-resonance spectrum.

If we truncate both Eqs. (12) and (13) keeping only the lowest averages, we obtain

$$\langle\rho_{22}(t)\rangle=\frac{1}{4}\frac{\Omega'^2}{K_2}\left(\frac{1}{2}\frac{\Omega^2}{K_1|R^0|^2/(R^0+R^{0*})+\Omega^2}\right)\times\frac{T^0+R^{0*}K_1/(R^0+R^{0*})}{S^0T^0+\frac{1}{4}\Omega^2}+\text{c.c.}\,, \tag{14}$$

which is the result obtained by the DA. Equation (14) also coincides with what is found for the PD model having the same spectrum as the CF, but is necessarily different in its higher-order correlations.[15] The DA obviously neglects terms of the order $(\Omega/b)^4$ and is, therefore, valid only for weak fields as we already noted before. The solution of Eq. (12) can be expressed in terms of an infinite sum over continued fractions. It turns out, however, to be much more convenient to solve Eq. (12) directly by numerical methods. Equation (12) is a band-structured linear algebraic equation which can be solved numerically very efficiently up to high orders since no pivoting is required. The convergence of $\langle\rho_{22}(t)\rangle=\rho_{22}^{00}$ depending on the truncation of the system can be checked easily since for $b=0$, where the convergence is worst, the exact solution (8) is known. In Figs. 1–4 we compare the result of numerical calcula-

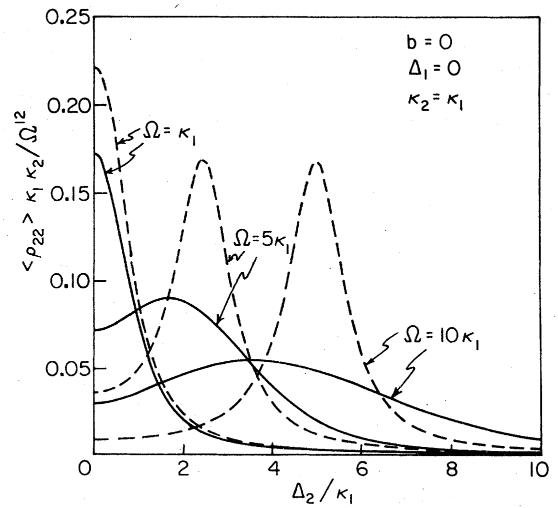

FIG. 1. Spectrum of double optical resonance. The exciting CF (solid lines) and coherent field (dashed lines) are monochromatic ($b=0$) and tuned on resonance ($\Delta_1=0$). The spectrum is symmetric around $\Delta_2=0$.

tions of $\langle \rho_{22}(t)\rangle$ for the CF (solid lines) with those for the phase diffusion model and DA equation (14) having the same spectrum and intensity as the CF.

Figures 1 and 2 show the population of level $|2\rangle$ as a function of the detuning of the probe laser Δ_2 for $\Delta_1=0$. The bandwidth is chosen as $b=0$ and $b=K_1$ in Figs. 1 and 2. In each of these figures the Rabi frequency is varied from $\Omega=K_1$, $5K_1$ to $10K_1$. Only the right half of the spectrum is shown since it is symmetric around $\Delta_2=0$. For low intensity corresponding to $\Omega=K_1$ and excita-

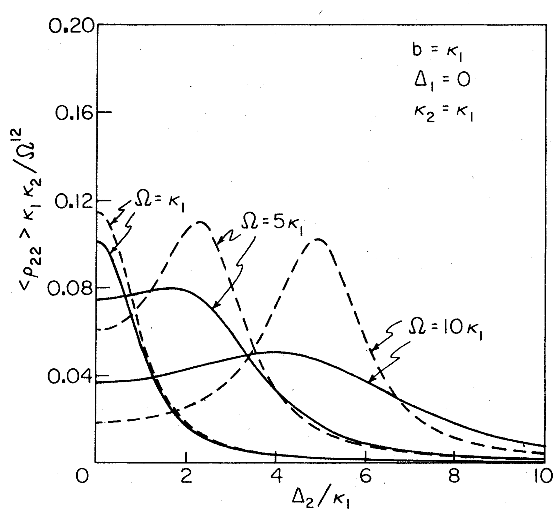

FIG. 2. Spectrum of double optical resonance. The exciting CF (solid lines) and phase-diffusing light (dashed lines) have finite bandwidth $b=K_1$ and are tuned on resonance ($\Delta_1=0$). The spectrum is symmetric around $\Delta_2=0$.

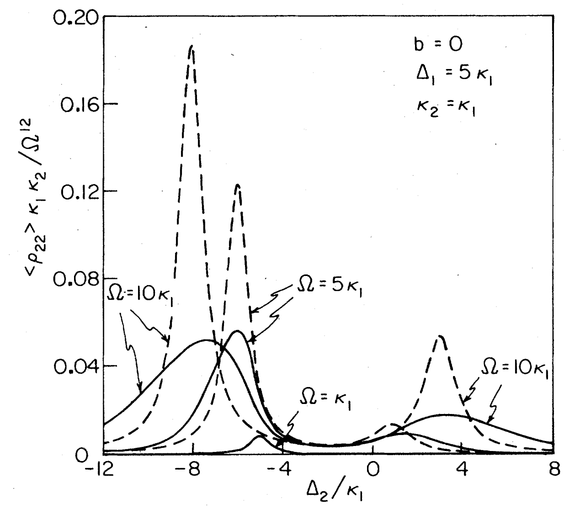

FIG. 3. Spectrum of double optical resonance. The exciting CF (solid lines) and coherent field (dashed lines) are monochromatic ($b=0$) and tuned off resonance ($\Delta_1=5K_1$).

tion by phase-diffusing radiation, the population of state $|2\rangle$ shows a single peak at the unperturbed transition frequency $|1\rangle-|2\rangle$ with linewidth determined essentially by the natural decay constants of the atom and bandwidth of the field. For the CF the intensity of this line is reduced since fewer atoms are excited to state $|1\rangle$.[19] Furthermore, the line shape in the CF is no longer Lorentzian but has broader wings. When the Rabi frequency is increased to $\Omega=5K_1$ and $10K_1$, saturating the transition $|0\rangle-|1\rangle$, the spectrum splits into a doublet. When the system $|0\rangle$, $|1\rangle$ is ex-

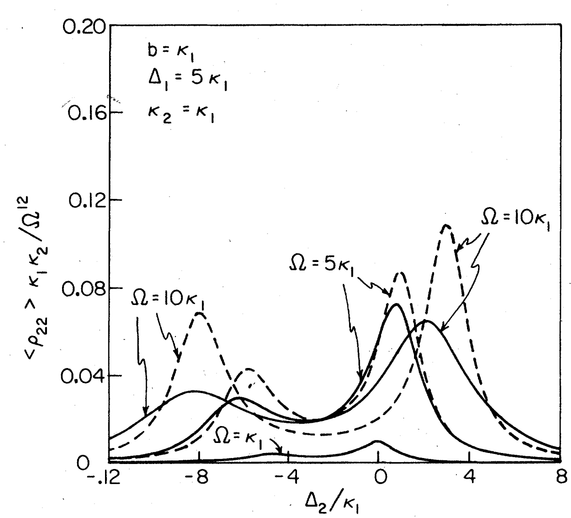

FIG. 4. Spectrum of double optical resonance. The exciting CF (solid lines) and phase-diffusing light (dashed lines) have finite bandwidth $b=K_1$ and are tuned off resonance ($\Delta_1=5K_1$).

cited by phase-diffusing light, the splitting frequency is approximately equal to the Rabi frequency Ω.[15] The heights and widths of these lines depend only on the natural decay constants but are practically independent of the intensity. The splitting in the CF may be understood qualitatively on the basis of Eq. (7) which states that for zero bandwidth fields $\langle\rho_{22}(t)\rangle$ is obtained by averaging the corresponding population in a coherent field $\rho_{22}^{b=0}(\epsilon,\epsilon^*)$ over the distribution $P_s(\epsilon,\epsilon^*)$. For high intensities $\rho_{22}^{b=0}(\epsilon,\epsilon^*)$ is proportional to two Lorentzians which, as we noted before, are shifted by $\pm\mu_{01}|\epsilon|$ relative to the unperturbed transition frequency. If in the average (8) these Lorentzians are replaced by two δ functions, thus neglecting broadening due to spontaneous decay, we find[27,28]

$$\langle\rho_{22}(t)\rangle=\int_0^\infty d|\epsilon|\,2\frac{|\epsilon|}{\langle|\epsilon|^2\rangle}\exp(-|\epsilon|^2/\langle|\epsilon|^2\rangle)\frac{1}{8}\frac{\Omega'^2}{K_2}\pi[\delta(\Delta_2-\mu_{01}|\epsilon|)+\delta(\Delta_2-\mu_{01}|\epsilon|)]=\frac{\Omega'^2}{K_2}\pi\frac{|\Delta_2|}{\Omega^2}e^{-4\Delta_2^2/\Omega^2},$$
$$|\Delta_2|,\Omega\gg K_1,K_2. \tag{15}$$

Equation (15) predicts a doublet structure with Rayleigh distribution for the line shape corresponding to the probability function of the amplitude of the complex CF. Note that in a real CF, which has a Gaussian amplitude distribution, no splitting would occur.[28] Equation (15) predicts a reduction of the splitting frequency to $(1/\sqrt{2})\Omega$ in agreement with Figs. 1 and 2. Contrary to the spectrum for the phase-diffusion model, a larger Rabi frequency now increases the width of the lines leading to a reduction of the heights. The total intensity is, however, approximately the same as for the PD model and is nearly independent of the intensity of the incident light. Although the maxima in the doublet decrease in proportion to $1/\Omega$, the ratio of the minimum at $\Delta_2=0$ to the maximum at $\Delta_2=\pm(1/2\sqrt{2})\Omega$ decreases for $b=0$ according to

$$\frac{\langle\rho_{22}\rangle_{\min}}{\langle\rho_{22}\rangle_{\max}}=\frac{\sqrt{2}\,e^{1/2}}{\pi}\frac{K_{02}+K_{01}}{\Omega}\left(\frac{K_{12}K_{02}\ln(\Omega^2/K_{12}K_{02})-\frac{1}{2}K_{01}K_1\ln(2\Omega^2/K_{01}K_1)}{K_{12}K_{02}-\frac{1}{2}K_{01}K_1}-0.577\right), \tag{16}$$

as may be found from Eq. (8), thus improving the visibility of the splitting for larger Rabi frequencies (Fig. 1). The doublet spectrum in the CF is insensitive to a finite bandwidth since for $\Omega\gg b$ the broadening of the lines due to the intensity fluctuations is dominant. Only the minimum at $\Delta_2=0$ is raised with increasing bandwidth (Fig. 2).

Figures 3 and 4 show the spectra for $b=0$ and $b=K_1$, respectively, when the field coupling $|0\rangle-|1\rangle$ is tuned off resonance with $\Delta_1=5K_1$. For $\Omega=K_1$ the spectra for phase-diffusing light and the CF are indistinguishable in these figures, as is expected from Eq. (14). The line at $\Delta_2\approx-\Delta_1$ corresponds to a nonresonant two-photon excitation $|0\rangle\overset{\omega}{\to}\overset{\omega'}{\to}|2\rangle$ which is proportional to the intensity of the strong field.[15] The peak at $\Delta_2\approx 0$ originates from the two-step process $|0\rangle\overset{\omega}{\to}|1\rangle\overset{\omega'}{\to}|2\rangle$ which for $b=0$ varies as the square of the intensity.[15] For finite bandwidth, photons in the tail of the Lorentzian spectrum of the exciting light, which are resonant with the transition $|0\rangle-|1\rangle$, pump this transition leading to a linear or weaker intensity dependence. For $b>K_1/2$ this two-step transition becomes even stronger than the two-photon excitation thus reversing the peak asymmetry in Fig. 4 in comparison with Fig. 3. Due to the long tail of the Lorentzian spectrum this reversed asymmetry persists for arbitrary detuning.[15] Since a realistic laser spectrum has wings falling faster than a Lorentzian, in an experiment this reversed asymmetry appears only for detunings not much larger than a few bandwidths of the field.[4] The reversed asymmetry, being a bandwidth effect, is also found in the CF spectrum. For $\Omega=5K_1$ and $10K_1$, the spectrum in the exciting phase-diffusing light shows peaks at approximately $\frac{1}{2}[-\Delta_1\pm(\Delta_1^2+\Omega^2)^{1/2}]$. A qualitative understanding of the different Stark shifts for the CF may be obtained by considering the limiting cases $\Omega\ll\Delta_1$ and $\Omega\gg\Delta_1$ in Eqs. (8) and (12). For $\Omega\ll\Delta_1$ the PD field induces a quadratic Stark shift of $-\frac{1}{2}\Omega^2/\Delta_1$ at the two-photon excitation line $\Delta_1+\Delta_2\approx 0$. Depending on whether $\frac{1}{2}\Omega^2/\Delta_1\gg K_{02}$, b or $\frac{1}{2}\Omega^2/\Delta_1\ll K_{02}$, b, the Stark shift in the CF of this line is found to be $-\frac{1}{2}\Omega^2/\Delta_1$ or $-2\frac{1}{2}\Omega^2/\Delta_1$, respectively. In the first case, where the fluctuations of the Stark shift, as induced by the intensity fluctuations of the CF, give the dominant contribution to the linewidth, the peaks of the lines in the PD and CF are shifted by the same amount (Fig. 3, $\Omega=5K_1$). In the second case, where the width is dominated by the natural linewidth or bandwidth of the field, the shift in the CF is enhanced by a factor of two (Fig. 4, $\Omega=5K_1$). This enhancement is related to recent findings in resonant multiphoton ionization in CF[17,30]: there it has been shown that in the limit of a Stark shift, small compared to the bandwidth of the CF, the shift is enhanced by a factor of $N+1$ for an N-photon resonance. If the intensity is increased, the broadening of the lines by the intensity fluctuations always finally dominates for $\Omega\gg K_1,K_2,b$. In this limit we find

$$\langle \rho_{22}(t)\rangle = \frac{\Omega'^2}{K_2}\,\pi\,\frac{2\Delta_2(\Delta_1+\Delta_2)}{\Delta_1^2+2\Delta_2(\Delta_1+\Delta_2)}\,\frac{|\Delta_2|}{\Omega^2} \times \exp^{-4\Delta_2(\Delta_1+\Delta_2)/\Omega^2}, \quad (\Delta_2(\Delta_1+\Delta_2>0). \tag{17}$$

Equation (17) predicts that for $\Omega \gg \Delta_1$ the peak of the two-photon excitation line occurs at $-\frac{3}{4}\Delta_1 - (1/2\sqrt{2})\Omega$, which means the peak in the CF is shifted less than for the PD model (Figs. 3 and 4, $\Omega = 10K_1$), as we found on resonance. The increased Stark shift for the two-step excitation line ($\Delta_2 \approx 0$) for $\Omega = 5K_1$ in Fig. 3 is explained by the quadratic intensity dependence of this line for $\Omega \ll \Delta_1$ and $b = 0$, giving rise to an enhancement of the Stark shift $\frac{1}{2}\Omega^2/\Delta_1$ by a factor of two or three for $\frac{1}{2}\Omega^2/\Delta_1 \gg K_{12}$ and $\frac{1}{2}\Omega^2/\Delta_1 \ll K_{12}$, respectively. In the case of a finite bandwidth CF, where the tail of the exciting Lorentzian spectrum populates the upper state $|1\rangle$, the intensity dependence of this line becomes linear or weaker.[15] To the lowest order in Ω/Δ_1 the excited state and therefore the two-step resonance line are shifted by the same amount for both the PD and the CF (Fig. 4, $\Omega = 5K_1$). According to Eq. (17), an increase of the intensity to $\Omega \gg \Delta_1$ results in a relative reduction of the Stark shift in the CF, in comparison with the PD model, to $-\frac{1}{4}\Delta_1 + (1/2\sqrt{2})\Omega$ (Fig. 4, $\Omega = 10K_1$).

The series of Figs. 1–4 clearly shows that the DA cannot even account qualitatively for the simplest features of the CF spectrum at high intensities indicating the inadequacy of this approach.

Up to now we confined our discussion to the case in which the transition $|0\rangle - |1\rangle$ is a one-photon resonance. For a two-photon resonance the Rabi frequency becomes proportional to the intensity. A qualitative understanding of the double-resonance spectrum at high intensities may be obtained in the approximation where the laser bandwidth and spontaneous decay are neglected. As in the derivation of Eqs. (15) and (17) on resonance we find

$$\langle \rho_{22}(t)\rangle = \frac{1}{4}\,\frac{\Omega'^2}{K_2\Omega}\, e^{-2|\Delta_2|\Omega}\,. \tag{18}$$

The spectrum is now an exponential function corresponding to the intensity distribution of the CF. Instead of the doublet splitting, only a broadened line on the transition frequency $|1\rangle - |2\rangle$ with a width proportional to the Rabi frequency appears. Similar results may be found for higher-order resonances. ac Stark splitting in double-resonance can therefore only occur in a CF, when a one-photon resonance is excited by a complex CF.

Before closing this section, we point out that the spectrum of fluorescence which may be observed from level $|1\rangle$ to an unperturbed level $|2\rangle$ with energy $\hbar\omega_2 < \hbar\omega_1$ obeys a system of equations nearly identical to Eq. (5).[29] If we identify $\Delta_2 = -(\omega_K - \omega_{12})$ with ω_K the frequency of the spontaneously emitted photon, Figs. 1–4 show the corresponding spectrum within a proportionality factor.[29]

IV. RESONANCE FLUORESCENCE

In this section we consider the spectrum of resonance fluorescence in a TLA $|0\rangle$, $|1\rangle$ driven by the intense nonmonochromatic CF of Sec. 2. The theory of resonance fluorescence for excitation by coherent radiation is well developed and in excellent agreement with experiment.[1,2,24-26] For a complete list of references on this subject we refer to Ref. 26. In order to generalize these theories so as to include the CF, we find it convenient to calculate the spectrum within an approach which for coherent radiation has been outlined by Mollow.[24] We separate the mode k of the radiation field from the state vector $|t\rangle$ of the atom-field system according to

$$|t\rangle = \sum_{\mu=0}^{1} |\mu\rangle|0\rangle_k |A_\mu(t)\rangle + \sum_{\mu=0}^{1} |\mu\rangle|1\rangle_k |B_{k\mu}(t)\rangle\,, \tag{19}$$

omitting higher photon numbers which give no contribution in the limit of infinite quantization volume.[24] From the expectation value of the number of photons in mode k,

$$N_k(t) = \sum_{\mu=0}^{1} \langle B_{k\mu}(t)|B_{k\mu}(t)\rangle\,,$$

the spectral emission rate of the TLA as the frequency ω_k is found to be[24]

$$\frac{d}{dt}N_k(t) = \vec{\mu}_{01}\vec{u}_k(\vec{x}=0)\langle A_1(t)|B_{k0}(t)\rangle + \text{c.c.}\,, \tag{20}$$

with $\vec{u}_k(\vec{x})$ the mode function of the radiation field. The quantities $\langle A_\mu(t)|B_{k\nu}(t)\rangle$ can be shown to be a solution of[24]

$$\left[i\frac{d}{dt} + \nu_k + \begin{pmatrix} \delta & 0 & -\mu_{01}\epsilon(t) & \mu_{01}\epsilon(t) \\ 0 & -\delta^* & \mu_{10}\epsilon^*(t) & -\mu_{10}\epsilon^*(t) \\ -\mu_{10}\epsilon^*(t) & \mu_{01}\epsilon(t) & iK_1 & 0 \\ \mu_{10}\epsilon^*(t) & -\mu_{01}\epsilon(t) & -iK_1 & 0 \end{pmatrix}\right] \begin{pmatrix} \langle A_0|B_{k1}\rangle \\ \langle A_1|B_{k0}\rangle \\ \langle A_1|B_{k1}\rangle \\ \langle A_0|B_{k0}\rangle \end{pmatrix} = i\vec{\mu}_{10}\vec{u}_k^*(\vec{x}=0) \begin{pmatrix} 0 \\ \rho_{11}(t) \\ 0 \\ \rho_{10}(t) \end{pmatrix}, \tag{21}$$

with $\delta=\Delta_1+\frac{1}{2}iK_1$ and $\nu_k=\omega-\omega_k$. In Eq. (21) the oscillations at the optical frequency have been removed according to the $\langle A_\mu|B_{k\nu}\rangle\to\langle A_\mu|B_{k\nu}\rangle e^{i(\mu-\nu-1)t}$. The terms on the right-hand side of Eq. (21) are the slowly varying density matrix elements of the TLA. The stationary solution of Eq. (21) for coherent zero-bandwidth fields, as has been given in Refs. 24–26, will be discussed below together with the spectrum in the CF.

When, owing to intensity and phase fluctuations, the incident field amplitude $\epsilon(t)$ becomes a stochastic function of time, the spectral emission rate $(d/dt)N_k(t)$ becomes a stochastically fluctuating quantity. As in the case of double resonance, these fluctuations are induced directly by the field as well as by the stochastic behavior of the density matrix elements. In order to obtain the averaged spectral emission rate $\langle\langle(d/dt)N_k(t)\rangle\rangle_t$, Eq. (21) must be solved for the average $\langle\langle A_1(t)|B_{k0}(t)\rangle\rangle_t$.

When $\epsilon(t)$ has only phase fluctuations according to the phase-diffusion model, Eq. (21) can be solved exactly.[10,14] The solution as obtained by the DA[11,12] coincides with the exact result,[10] due to the fact that phase diffusion is a process with independent increments.[15] As a result of averaging over the phase, new damping terms proportional to the laser linewidth b appear in Eq. (21) (see Eq. (7) of Ref. 14). A discussion of the modification of the spectrum by the finite bandwidth will be given below.

For the CF the DA is inadequate in determining the spectrum; instead Eq. (21) must be solved by the method employed in Sec. III. In the zero-bandwidth limit the average may be performed by averaging the spectrum in the coherent field [see Eq. (95) of Ref. 26.] over the stationary field distribution $P_S(\epsilon,\epsilon^*)$ of the CF. In this way we obtain

$$\left\langle\frac{d}{dt}N_k(t)\right\rangle^{b=0}=|\vec{\mu}_{01}\vec{u}_k|^2\left\{\pi a[(1+a)e^aE_1(a)-1]\,\delta(\nu_k)+\frac{1}{4}\frac{K_1}{\nu_k^2+(K_1/2)^2}\times\left[1+\frac{a^2(c-a)}{|a-b|^2}e^aE_1(a)+\left(\frac{b^2(c-b)}{(b-b^*)(b-a)}e^bE_1(b)+\text{c.c.}\right)\right]\right\},\tag{22}$$

with

$$a=2|\delta|^2/\Omega^2,\quad b=-(\nu_k+iK_1)(\nu_k+\delta)(\nu_k-\delta^*)/[\Omega^2(\nu_k+iK_1/2)]\,,\quad c=2(\nu_k^2+K_1^2)/\Omega^2\,,$$

and E_1 the exponential integral.[33] The first term corresponds to the averaged coherent spectrum, while the other contributions come from the incoherent part of the spectral emission rate.[34] For finite bandwidth, Eq. (21) can be solved in a way similar to that employed for Eq. (5) encountered in double resonance. Defining $\langle A_\mu|B_{k\nu}\rangle^{\alpha n}$ in analogy to Eq. (9), we get from Eq. (A8) of the Appendix:

$$\left[i\frac{d}{dt}+\nu_k+\delta+i2(n+1)b\right]\langle A_0|B_{k1}\rangle^{-2n}-\tfrac{1}{2}\Omega\sqrt{n+2}(\langle A_1|B_{k1}\rangle^{-1n}-\langle A_0|B_{k0}\rangle^{-1n})+\tfrac{1}{2}\Omega\sqrt{n+1}(\langle A_1|B_{k1}\rangle^{-1n+1}-\langle A_0|B_{k0}\rangle^{-1n+1})=0\,,$$

$$\left[i\frac{d}{dt}+\nu_k-\delta^*+i2nb\right]\langle A_1|B_0\rangle^{0n}+\tfrac{1}{2}\Omega\sqrt{n+1}(\langle A_1|B_{k1}\rangle^{-1n}-\langle A_0|B_{k0}\rangle^{-1n})-\tfrac{1}{2}\Omega\sqrt{n}(\langle A_1|B_{k1}\rangle^{-1n-1}-\langle A_0|B_{k0}\rangle^{-1n-1})=i\vec{\mu}_{01}\vec{u}_k^*\rho_{11}^{0n}(t)\,,\tag{23}$$

$$\left[i\frac{d}{dt}+\nu_k+iK_1+i(2n+1)b\right]\langle A_1|B_{k1}\rangle^{-1n}-\tfrac{1}{2}\Omega(\sqrt{n+2}\langle A_0|B_{k1}\rangle^{-2n}-\sqrt{n}\langle A_0|B_{k1}\rangle^{-2n-1})+\tfrac{1}{2}\Omega\sqrt{n+1}(\langle A_1|B_{k0}\rangle^{0n}-\langle A_1|B_{k0}\rangle^{0n+1})=0$$

$$\left[i\frac{d}{dt}+\nu_k+i(2n+1)b\right]\langle A_0|B_{k0}\rangle^{-1n}-iK_1\langle A_1|B_{k1}\rangle^{-1n}+\tfrac{1}{2}\Omega(\sqrt{n+2}\langle A_0|B_{k1}\rangle^{-2n}-\sqrt{n}\langle A_0|B_{k1}\rangle^{-2n-1})+\tfrac{1}{2}\Omega\sqrt{n+1}(\langle A_1|B_{k0}\rangle^{0n}-\langle A_1|B_{k0}\rangle^{0n+1})=i\vec{\mu}_{01}\vec{u}_k^*\rho_{10}^{-1n}(t)\,,\quad n=0,1,\ldots\,.$$

In the stationary limit the spectral emission rate is then given by

$$\left\langle\frac{d}{dt}N_k(t)\right\rangle=\vec{\mu}_{01}\vec{u}_k(-\tfrac{1}{2}\Omega D^0+i\vec{\mu}_{01}\vec{u}_k^*\rho_{11}^{00})/(\nu_k-\delta^*)+\text{c.c.}\,,\tag{24}$$

where $D^n=\langle A_1|B_{k1}\rangle^{-1n}-\langle A_0|B_{k0}\rangle^{-1n}$ obeys the inhomogeneous three-term recursion formula

$$\left(\nu_k+i(2n+1)b+iK_1-\frac{(n+2)\frac{1}{2}\Omega^2}{\nu_k+\delta+i2(n+1)b}-\frac{n\frac{1}{2}\Omega^2}{\nu_k+\delta+i2nb}-\frac{(n+1)\frac{1}{2}\Omega^2}{\nu_k-\delta^*+i2nb}-\frac{(n+1)\frac{1}{2}\Omega^2}{\nu_k-\delta^*+i2(n+1)b}\right)D^n$$
$$+\tfrac{1}{2}\Omega^2\sqrt{n(n+1)}\left(\frac{1}{\nu_k+\delta+i2nb}+\frac{1}{\nu_k-\delta^*+i2nb}\right)D^{n-1}+\tfrac{1}{2}\Omega^2\sqrt{(n+1)(n+2)}$$
$$\times\left(\frac{1}{\nu_k+\delta+i2(n+1)b}+\frac{1}{\nu_k-\delta^*+i2(n+1)b}\right)D^{n+1}$$
$$=i\vec{\mu}_{01}\vec{u}^{\,*}_k\left(-\frac{\sqrt{n+1}\,\Omega}{\nu_k-\delta^*+i2nb}\,\rho_{11}^{0n}+\frac{\sqrt{n+1}\,\Omega}{\nu_k-\delta^*+i2(n+1)b}\,\rho_{11}^{0n+1}-\frac{\nu_k+iK_1+(2n+1)ib}{\nu_k+i(2n+1)b}\,\rho_{10}^{-1n}\right) \tag{25}$$

and the averaged density matrix elements ρ_{11}^{0n} and ρ_{10}^{-1n} are solutions of Eq. (10). Equation (25) has an analogous interpretation as the corresponding equation (10) in double resonance: the coupling of D^0 to all higher-order averages D^n demonstrates the effect of all higher-order field correlations on the spectrum. The inhomogenous terms are due to fluctuations in the density matrix elements of the system $|0\rangle$, $|1\rangle$. If Eq. (25) is truncated in lowest order, we recover the decorrelation result which already differs from the corresponding solution for the PD model since even in the lowest approximation information on the second-order field correlation function enters.

We have solved Eq. (25) numerically. Figs. 5–8 compare the results of these calculations for the CF (solid lines) with those for the PD model (dashed lines). For $b=0$ only the incoherent part of the spectrum is shown in these figures.

Figures 5 and 6 give a graphical representation of the on-resonance spectrum ($\Delta_1=0$) for $b=0$ and $b=K_1$, respectively. In each figure the Rabi frequency assumes the values $\Omega=25K_1$ and $50K_1$. The spectrum is symmetric around $\omega_k=\omega$. The left-hand side of the spectrum is plotted on a linear scale while for $\omega_k \geq \omega$ a logarithmic scale is chosen. For monochromatic coherent excitation Fig. 5 shows the well-known triplet with central line at the laser frequency and sidebands shifted by the Rabi frequency.[24-26] The central-to-side peak ratio is 3:1 while the corresponding ratio of the widths is 1:3/2, in excellent agreement with experiments.[1,2] In finite bandwidth phase-diffusing light these ratios are modified to $(K_1+4b)^{-1}$: $\frac{1}{2}(\frac{3}{2}K_1+3b)^{-1}$ and $(K_1+4b):(\frac{3}{2}K_1+3b)$, respectively (Fig. 6).[26,35] For the CF a triplet spectrum is also observed. The existence of ac Stark splitting in resonance fluorescence for a CF can be understood qualitatively for $b=0$ by averaging the incoherent triplet spectrum under coherent excitation over the CF distribution $P_s(\epsilon,\epsilon^*)$ neglecting the broadening of the sidebands by spontaneous decay:

$$\left\langle\frac{d}{dt}N_k(t)\right\rangle^{b=0} = \frac{1}{2}\,|\vec{\mu}_{01}\mu_k|^2\left(\frac{\frac{1}{2}K_1}{\nu_k^2+(\frac{1}{2}K_1)^2}+\pi\frac{|\nu_k|}{\Omega^2}\,e^{-\nu_k^2/\Omega^2}\right) \quad (\Omega \gg K_1). \tag{26}$$

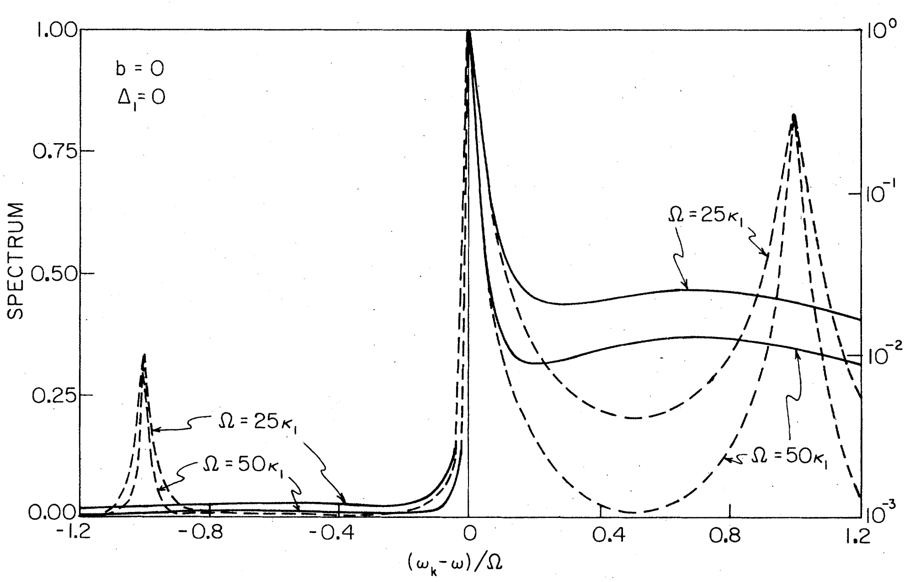

FIG. 5. Spectrum of resonance fluorescence. The exciting CF (solid lines) and coherent field are monochromatic ($b=0$) and tuned on resonance ($\Delta_1=0$). The spectrum is symmetric around $\omega_k=\omega$. The left- and right-hand sides of the spectrum are plotted on a linear and logarithmic scale, respectively. The coherent δ-function contribution at $\omega_k=\omega$ has been omitted.

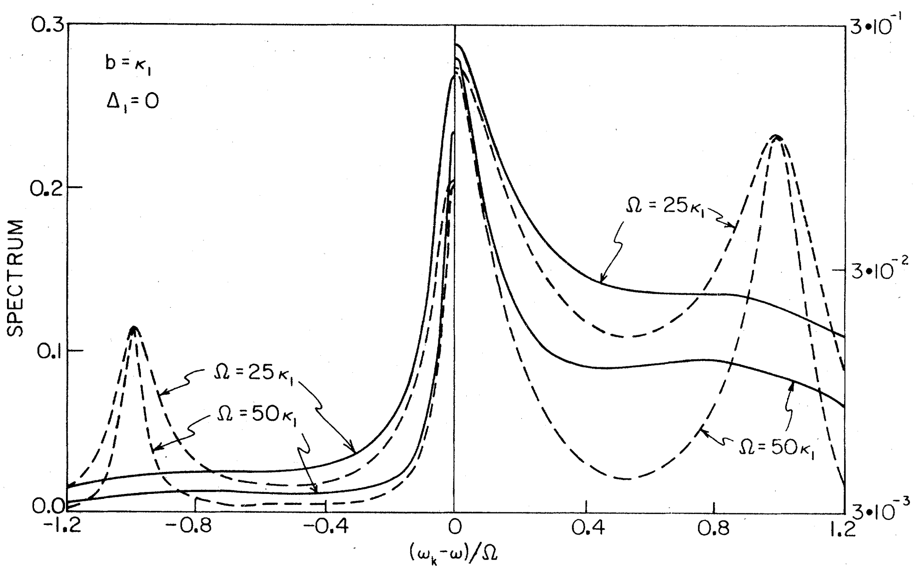

FIG. 6. Spectrum of resonance fluorescence. The exciting CF (solid lines) and phase-diffusing light (dashed lines) have finite bandwidth ($b = K_1$) and are tuned on resonance ($\Delta_1 = 0$). The spectrum is symmetric around $\omega_k = \omega$. The left-and right-hand sides of the spectrum are plotted on a linear and logarithmic scale, respectively.

The central line is unaffected by the intensity fluctuations, while the sidebands are smeared out according to the Rayleigh distribution of the CF amplitude, predicting a splitting frequency of $(1/\sqrt{2})\Omega$. Since the wings of the central Lorentzian tend to hide the sideband structure, the triplet spectrum only appears for $\Omega \geq 10K_1(b=0)$. For these large values of Ω the intensity of the sidebands is already very small: the central-to-side peak ratio is $2/K_1 : \pi/\sqrt{2e}\Omega$ which increases with Ω. Although the sideband maxima go down with $1/\Omega$, the ratio of the minimum between the mainline and the sideband to the sideband maximum decreases as $(9e/2)^{1/2}\,(K_1/\pi\Omega)^{1/3}$ thus improving the relative resolution of the triplet structure (Fig. 5). A finite bandwidth of the CF broadens the central line (Fig. 6). The broadening is always less than in the phase diffusion model since the decay constants of the Nth-order two-time correlation function of the CF increase only with Nb, compared to N^2b for phase-diffusing light.[5] Since the broadened central line has an increased overlap with the sidebands, the finite bandwidth raises the threshold for splitting to higher Rabi frequencies.

It is interesting at this point to compare our re-

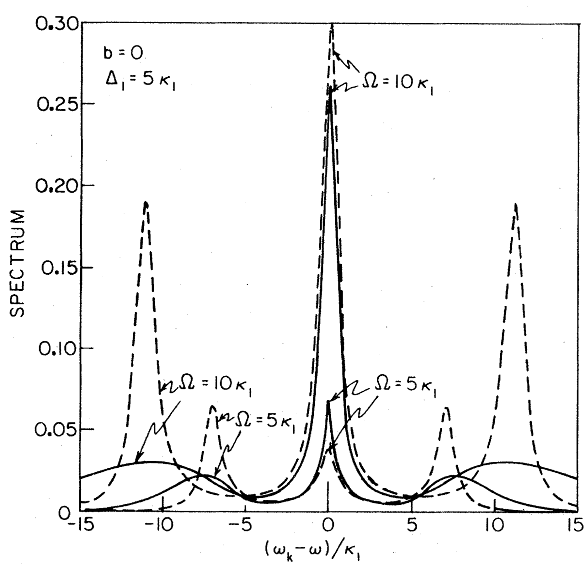

FIG. 7. Spectrum of resonance fluorescence. The exciting CF (solid lines) and coherent field are monochromatic ($b=0$) and tuned off resonance. The coherent δ-function contribution at $\omega_k = \omega$ has been omitted.

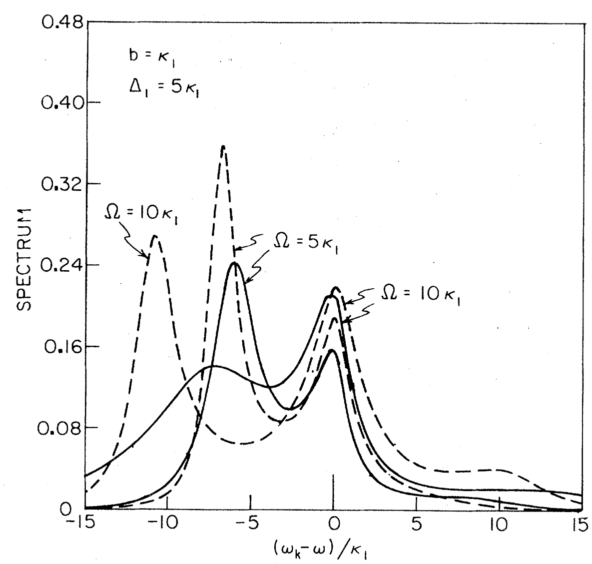

FIG. 8. Spectrum of resonance fluorescence. The exciting CF (solid lines) and phase-diffusing light (dashed lines) have finite bandwidth ($b = K_1$) and are tuned off resonance ($\Delta_1 = 5K_1$).

sults with those of Avan and Cohen-Tannoudji,[13] and Eberly.[11] The suppression of the sidebands of the triplet in resonance fluorescence had been anticipated qualitatively by Avan and Cohen-Tannoudji, although the triplet structure is not completely washed out as their arguments had suggested. Eberly's predictions regarding the effect of amplitude fluctuations in resonance fluorescence are based on the DA, which, in Eberly's form, only retains information about the lowest-order field correlation function. He decouples phase and amplitude fluctuations which can be justified for a single-mode laser far above threshold having small intensity fluctuations around a constant intensity.[36] On the other hand, he assumes the constant part of the intensity to be zero. Consequently, Eberly's conclusion regarding the modification of the heights and widths of the triplet cannot be compared with our results.[36]

Figures 7 and 8 show the off-resonance spectrum with $\Delta_1 = 5K_1$ for $b=0$ and $b=K_1$, respectively. Ω takes on the values $5K_1$ and $10K_1$. With excitation by monochromatic coherent radiation the incoherent spectrum consists of two symmetric sidebands separated by $(\Delta_1^2+\Omega^2)^{1/2}$ from the central line at the laser frequency (Fig. 7).[34] The dominant contribution to the spectrum at $\omega_k=\omega$ comes from the coherent δ function.[34] A finite bandwidth according to the PD model makes the spectrum asymmetric (Fig. 8).[12,35,37] The sideband nearer to the atomic transition frequency ω_{10} is enhanced since photons in the tail of the Lorentzian spectrum are resonant with ω_{10} and favor the two-step process $|0\rangle \xrightarrow{\omega} |1\rangle \xrightarrow{\omega_k} |0\rangle$. At the same time the other sideband is broadened and decreases in intensity. In the monochromatic CF the sidebands are still symmetric (Fig. 7). For $\Omega = 5K_1$ the peaks of these sidebands are shifted outwards compared to those in the coherent field. This increased splitting frequency for $b=0$ is explained by the quadratic intensity dependence[6] of the sidebands for $\Omega \ll \Delta_1$ in the coherent excitation spectrum: depending on whether the fluctuations in the Stark shift or the spontaneous decay width dominate the broadening of the sidebands, the Stark shift in the CF is enhanced by a factor of two or three in analogy to our discussion of double optical resonance in Sec. III. With increasing intensities these peaks are shifted inwards as we found on resonance (Fig. 7, $\Omega = 10K_1$). The incoherent line at $\omega_k=\omega$ is more pronounced in the CF as is again explained by the quadratic intensity dependence of this line for $\Omega \ll \Delta_1$ leading to a 2! enhancement for the CF (Fig. 7, $\Omega = 5K_1$). The coherent δ function of Eq. (22), which cannot be graphically represented in Fig. 7, gives the dominant contribution to the spectrum at $\omega_k=\omega$. For the monochromatic CF this coherent line is less intense than for the coherent field since fewer atoms are excited by the CF.[19] In the finite bandwidth case the sideband nearer to the atomic transition frequency is again enhanced (Fig. 8). Due to the intensity fluctuations this line is always broader than for the PD model. Contrary to the spectrum for $b=0$, this sideband is always shifted less than for the PD field since for $\Omega \ll \Delta_1$ the spontaneous emission $|1\rangle \xrightarrow{\omega_k} |0\rangle$ of the second step of the two-step process occurs at the atomic transition frequency which to the lowest order in Ω/Δ_1, is shifted by the same amount for both the CF and the PD model. The other sideband, which is suppressed in the finite bandwidth field, shows an enhanced Stark shift for the CF which is again explained by the quadratic intensity dependence[35] of this line for $\Omega \ll \Delta_1$. The central line in Fig. 8 consists of the coherent and incoherent contribution of the spectrum. It is dominated by the coherent part of the spectrum, having a linewidth determined by the spectral width of the exciting light. For the CF this line is again weaker than for the phase-diffusing light since fewer atoms are excited by the CF.[19]

V. CONCLUSIONS

We have investigated the splitting of an atomic transition in an intense nonmonochromatic chaotic driving field. We have shown that in both double optical resonance and resonance fluorescence, Stark splitting in a complex CF may be observed, although in resonance fluorescence the central line of the triplet spectrum tends to hide the sideband structure. On resonance and for high intensities the splitting frequency is reduced in comparison to coherent excitation. In this high-intensity limit the spectral line shape of the lines in double optical resonance and of the sidebands in resonance fluorescence reflect the amplitude distribution of the exciting light. With increasing intensity these lines broaden, which can be understood in terms of the intensity fluctuations inducing fluctuations in the Rabi frequency which tend to supress the lines in the doublet spectrum and the sidebands in resonance fluorescence. Except for the broadening of the central line in resonance fluorescence, at high intensities the finite bandwidth of the exciting light has only a minor effect: it is the intensity fluctuations of the CF which determine the spectrum. Off resonance we have found that for fields of finite bandwidth the asymmetry in double optical resonance reverses and a sideband asymmetry in resonance fluorescence appears. Depending on the relative magnitude of the parameters, the Stark shift (and therefore the splitting frequency) may be larger or smaller than for coherent excitation.

A chaotic field has thus been shown to introduce significant modifications in the saturation behavior of a TLA. There seems to exist some evidence that similar effects have been seen in double-resonance experiments.[4] It is hoped that future experimental investigations in double optical resonance, as well as resonance fluorescence, will provide further insight into these effects.

ACKNOWLEDGMENTS

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APPENDIX

The stochastic differential equations (5), (6), and (21) are of the form

$$\dot{X}_\mu(t)=\sum_\nu A_{\mu\nu}(\epsilon(t),\epsilon^*(t))X_\nu(t)\,, \qquad (A1)$$

where in Sec. III $X_\mu(t)$ is a density-matrix element or, as in Sec. IV, a variable describing the spectral emission rate of the TLA. $A_{\mu\nu}(\epsilon(t),\epsilon^*(t))$ is a matrix depending on the stochastic driving field $\epsilon(t)$ which determines the time evolution of the system. Equation (A1) must be solved for the averages $\langle X_\mu(t)\rangle_t$ where $\langle\ \rangle_t$ denotes averaging of $X_\mu(t)$ with respect to the fluctuations in the driving field. The subscript t indicates the explicit time dependence of the average. Since $\epsilon(t)$ obeys a Markov process, as we assumed for our CF so that the corresponding distribution function $P(\epsilon,\epsilon^*,t)$ obeys a master equation of the form

$$[\partial/\partial t+L(\epsilon,\epsilon^*)]P(\epsilon,\epsilon^*,t)=0\,,$$

the averages $\langle X_\mu(t)\rangle_t$ may be found by solving the system of equations[32]

$$\left[\frac{\partial}{\partial t}+L(\epsilon,\epsilon^*)\right]X_\mu(\epsilon,\epsilon^*,t)=\sum_\nu A_{\mu\nu}(\epsilon,\epsilon^*)X_\nu(\epsilon,\epsilon^*,t) \qquad (A2)$$

and integrating the solution according to

$$\langle X_\mu(t)\rangle_t=\int d^2\epsilon X_\mu(\epsilon,\epsilon^*,t)\,. \qquad (A3)$$

Since for our model L is given by (3), Eq. (A2) is a system of partial differential equations which must be solved under the initial condition

$$X_\mu(\epsilon,\epsilon^*,t=0)=\langle X_\mu(t)\rangle_{t=0}P_s(\epsilon,\epsilon^*) \qquad (A4)$$

with $P_s(\epsilon,\epsilon^*)$ the stationary distribution function.[31] Equation (A2) can be solved by expanding $X_\mu(\epsilon,\epsilon^*,t)$ in the complete biorthogonal set of eigenfunctions of L[30]:

$$LP_{\alpha n}(\epsilon,\epsilon^*)=\Lambda_{\alpha n}P_{\alpha n}(\epsilon,\epsilon^*)\,,$$
$$L^\dagger\phi_{\alpha n}(\epsilon,\epsilon^*)=\Lambda^*_{\alpha n}\phi_{\alpha n}(\epsilon,\epsilon^*)\,. \qquad (A5)$$

For the CF of Sec. II these eigenfunctions have been shown to be given by[30]

$$P_{\alpha n}(\epsilon,\epsilon^*)=P_s(\epsilon,\epsilon^*)\phi_{\alpha n}(\epsilon,\epsilon^*),\ \alpha=0,\pm1,\ldots,\quad n=0,1,2,\ldots,$$

$$\phi_{\alpha n}(\epsilon,\epsilon^*)=\left(\frac{n!}{(n+|\alpha|)!}\right)^{1/2}|\epsilon|^{|\alpha|}(\epsilon^*/\epsilon)^{\alpha/2}\times L_n^{|\alpha|}(|\epsilon|^2/\langle|\epsilon|^2\rangle)/\langle|\epsilon^2|\rangle^{|\alpha|/2}, \qquad (A6)$$

with eigenvalues $\Lambda_{\alpha n}=b(2n+|\alpha|)$. With the definition

$$X_\mu^{\alpha n}(t)=\int d^2\epsilon\phi^*_{\alpha n}(\epsilon,\epsilon^*)X_\mu(\epsilon,\epsilon^*,t)=\langle\phi^*_{\alpha n}(\epsilon(t),\epsilon^*(t))X_\mu(t)\rangle_t\,, \qquad (A7)$$

we find from Eq. (A2) an infinite system of differential equations for the one-time atom-field averages (A7),

$$\left(\frac{d}{dt}+\Lambda_{\alpha n}\right)X_\mu^{\alpha n}(t)=\sum_{\beta m\nu}\int d^2\epsilon\phi^*_{\alpha n}(\epsilon,\epsilon^*)A_{\mu\nu}(\epsilon,\epsilon^*)P_{\beta m}(\epsilon,\epsilon^*)\times X_\nu^{\beta m}(t)\,. \qquad (A8)$$

Explicit expressions for these matrix elements have been given in Ref. 30. Note that due to $\phi_{00}=1$ the averages $\langle X_\mu(t)\rangle_t$ are simply given by $\langle X_\mu(t)\rangle_t=X_\mu^{00}(t)$.

In the stationary limit, when all transients have died out, Eq. (A8) reduces to an infinite set of algebraic equations for $\langle X_\mu(t)\rangle$. Although these averages become time independent in this stationary limit, the $X_\mu(t)$'s are not, and we are not able to neglect the time derivative in Eq. (A1). Applying the above formalism to Eqs. (5), (6), and (21), as encountered in Secs. III and IV, we find with the help of Eq. (A8) that Eqs. (5), (6), and (21) reduce to Eqs. (10) and (23), respectively.

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