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Saturation and Stark splitting of an atomic transition in a stochastic field

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The authors investigate the saturation and Stark splitting of an atomic transition in (i) a phase-diffusion field and (ii) a chaotic field of arbitrary bandwidth. The theory takes into account the infinite sequence of field-correlation functions. It is shown that a chaotic field is less effective than a phase-diffusion field in saturating a single- or multiphoton transition. This is contrary to the weak-field case, where the intensity fluctuations and the associated photon bunching make the chaotic field more effective in exciting a multiphoton transition. It is also shown that the Stark splitting of an atomic transition, as observed in double resonance, is influenced dramatically by the intensity fluctuations in the chaotic field.

I. INTRODUCTION

Most of the theoretical work on the resonant interaction of strong electromagnetic radiation with matter has been based on the assumption of monochromatic and purely coherent radiation. Even lasers, however, the most coherent sources of optical radiation, do not always satisfy this assumption as their bandwidth and departure from coherence are not necessarily negligible. In single-mode operation and at the expense of reduced power, the laser bandwidth can indeed be very small. Many experiments, however, are performed with high-power multimode laser pulses whose bandwidth is much larger than typical atomic linewidths and whose intensity undergoes substantial fluctuations. Basically, real lasers undergo intensity, phase, and frequency fluctuations to varying degrees, and depending on the experimental circumstances, should be treated properly as stochastic processes. Numerous papers[1] and several books[2] have dealt with the classical as well as the quantum theory of coherence and the statistical properties of radiation. Considerable attention has also been given to the theory of the interaction of radiation with matter generalized so as to include the statistical properties of the radiation. Mathematically, this generalization is simple only for linear processes (weak fields) and nonresonant multiphoton (nonlinear) processes. The mathematical treatment of the more interesting case of nonlinear resonant processes in strong, stochastically fluctuating fields, is generally very difficult.[3-17]

Such resonant processes can be formulated in terms of an atomic density matrix $\rho(t)$ coupled to a stochastic field of amplitude $\epsilon(t)$. The equation of motion of $\rho(t)$ must then be averaged over the field fluctuations. But for N-photon resonance, this averaging leads to atomic-field correlation functions of the type $\langle\epsilon^{*N}(t_1)\epsilon^{N}(t_2)\rho_{ii}(t_2)\rangle$. Generally, such a correlation cannot be evaluated without first solving the stochastic differential equation for $\rho(t)$, which in general is a horrendous task. The decorrelation

$$\langle\epsilon^{*N}(t_1)\epsilon^{N}(t_2)\rho_{ii}(t_2)\rangle = \langle\epsilon^{*N}(t_1)\epsilon^{N}(t_2)\rangle\langle\rho_{ii}(t_2)\rangle\ ,$$

which was first used in this context by Apanasevich *et al.*[3] ($N=1$), is valid only for Wiener-Levy-type phase fluctuations. In the case of a general stochastic field, this decorrelation can be used as a first approximation only for weak fields below saturation. For strong fields the decorrelation approximation can lead to erroneous predictions. One of these erroneous predictions, for example, is that a chaotic field is always more effective than a coherent field in saturating an N-photon transition. Recall that for weak fields, N-photon absorption in a chaotic field is enhanced by a factor of $N!$ relative in that in a coherent field. In the decorrelation approximation, as the intensity increases and the transition saturates, this enhancement is predicted to decrease monotonically to unity. This prediction is, however, incorrect. Actually, as we show in this paper with increasing intensity the enhancement decreases rapidly to a minimum value below unity, and then goes to unity. Therefore, the chaotic field is less effective than a coherent field, in saturating an N-photon transition. This is one of the new results in this paper. The incorrect prediction of the decorrelation approximation is caused by the fact that in the decorrelation approximation the equations describing the interaction contain information only about the Nth-order field correlation function. For weak fields this information is adequate, because the average N-photon absorption depends only on the Nth-order field correlation. For strong fields, however, because of saturation the average N-photon absorption depends on the infinite sequence of field correlations of order N^K, $K=1,2,\ldots$. Therefore, as we have pointed out before,[16] any theory based on the decorrelation approximation, for a general field, is inherently a weak-field theory. As we show in this paper, in the presence of intensity fluctuations, whether they are chaotic or not, the decorrelation is not valid. Consequently, the predictions of Ref. 9 on the effect of intensity fluctuations on the spectrum of resonance fluorescence, based on the decorrelation approximation, are incorrect. Specifying only the first-order field correlation simply is not sufficient to solve the problem of resonance fluorescence in the presence of intensity fluctuations.[4]

In this paper we investigate the saturation and Stark splitting of an atomic transition in an intense fluctuating field with arbitrary bandwidth. The field is treated as a classical stochastic process described statistically by specifying the infinite sequence of the field-correlation functions. In the absence of such complete statistical information about real laser fields, we consider two well-known models for the stochastic field (a) the phase-diffusion (PD) field (Wiener-Levy-type phase fluctuations) and (b) the chaotic (CH) field. For the phase-diffusion model the decorrelation of the atomic and field variables is rigorous. This results from the statistical independence of the phase increments for Wiener-Levy-type phase fluctuations. For the chaotic field, the decorrelation is not valid. For a chaotic field of zero bandwidth, we find the exact correction to the decorrelation approximation. For a chaotic field of nonzero bandwidth, we obtain a perturbation series expansion for the correction to the decorrelation approximation. In the case of a Markovian chaotic field, this perturbation series is summed to all orders.

II. SATURATION IN A TWO-LEVEL SYSTEM

We consider a two-level atom with ground state $|1\rangle$ and excited state $|2\rangle$. The matrix element of the electric dipole between the two states is μ_{12} and the transition frequency ω_{21}. The system is interacting with a fluctuating electric field

$$E(t)=\epsilon(t)e^{i\omega t}+\epsilon^*(t)e^{-i\omega t}\,, \tag{1}$$

where ω is the center frequency of the spectrum and $\epsilon(t)\equiv|\epsilon(t)|e^{i\phi(t)}$ is the fluctuating complex amplitude, with $|\epsilon(t)|$ and $\phi(t)$ being the real amplitude and phase, respectively. The fluctuating complex amplitude $\epsilon(t)$ is treated here as a stochastic process. In this paper we consider two well-known models for the stochastic field: (a) the phase-diffusion field and (b) the chaotic field.

The phase-diffusion field has a constant amplitude but its phase is a Wiener-Levy stochastic process (Brownian motion with negligible acceleration).[18] A Wiener-Levy process is a nonstationary Markov Gaussian process whose increments are independent, i.e.,

$$\langle[\phi(t_1)-\phi(t_2)][\phi(t_2)-\phi(t_3)]\rangle$$
$$=\langle\phi(t_1)-\phi(t_2)\rangle\langle\phi(t_2)-\phi(t_3)\rangle\,,\quad t_1>t_2>t_3\,,$$

where the angular brackets denote stochastic average. The complex amplitude of the phase-diffusion field has zero mean value $[\langle\epsilon(t)\rangle=0]$ and its nth-order correlation function is given by[2]

$$\langle\epsilon^*(t_1)\epsilon(t_2)\cdots\epsilon^*(t_{2n-1})\epsilon(t_{2n})\rangle$$
$$=\prod_{j\text{ odd}}^{2n-1}\langle\epsilon^*(t_j)\epsilon(t_{j+1})\rangle\,, \tag{2}$$

where $t_1>t_2\cdots t_{2n-1}>t_{2n}$. Since $\epsilon(t)$ is a stationary Markov process, the first-order correlation function is necessarily exponential,[18] i.e.,

$$\langle\epsilon^*(t_1)\epsilon(t_2)\rangle=\epsilon_0^2\exp(-\tfrac{1}{2}\gamma|t_1-t_2|)\,, \tag{3}$$

where γ is the full width at half-maximum (FWHM) of the Lorentzian spectrum and $\epsilon_0^2=\langle|\epsilon(t)|^2\rangle$ is the variance of $\epsilon(t)$. Note that $\langle\phi(t_1)\phi(t_2)\rangle=\gamma\tau$, where τ is the smallest of the two times t_1 and t_2, and $\langle\dot\phi(t_1)\dot\phi(t_2)\rangle=\gamma\delta(t_1-t_2)$, where the dot denotes time derivative.

The chaotic field is a complex Gaussian stochastic process, with both amplitude and phase fluctuations. It can be written as $\epsilon(t)=\epsilon_x(t)+i\epsilon_y(t)$, where $\epsilon_x(t)$ and $\epsilon_y(t)$ are two independent real Gaussian stochastic processes with zero mean value and equal variance. The complex amplitude of the chaotic field has also zero mean value and its nth-order correlation function is given by[2]

$$\langle\epsilon^*(t_1)\epsilon(t_2)\cdots\epsilon^*(t_{2n-1})\epsilon(t_{2n})\rangle$$
$$=\sum_P\prod_{j\text{ odd}}^{2n-1}\langle\epsilon^*(t_j)\epsilon[t_{P(j+1)}]\rangle\,, \tag{4}$$

where $t_1>t_2>\cdots>t_{2n-1}>t_{2n}$ and P denotes permutation. A complex Gaussian stochastic process, such as the chaotic field, is not necessarily Markovian and hence its spectrum is not necessarily Lorentzian. However, because of simplifications in the calculations later on, and for the purpose of comparison with the phase-diffusion field, we will assume that the chaotic field is also Markovian and that its first-order correlation function is given by Eq. (3). Note that the phase-diffusion field corresponds to an intensity stabilized single-mode laser field. The chaotic field, on the other hand, corresponds to a multimode laser field with a large number of uncorrelated modes. A chaotic field can also be synthesized by passing a single-mode laser beam through a rotating ground-glass disk.

The bandwidth of such a chaotic field is proportional to the rotation frequency of the disk.[19]

The equation of motion for the density matrix $\rho(t)$ of a two-level system, in the rotating-wave approximation [i.e., $\rho_{12}(t)=\sigma_{12}(t)e^{i\omega t}, \rho_{ii}(t) = \sigma_{ii}(t)$, $i=1,2$, where the $\sigma_{ij}(t)$ are slowly varying amplitudes] can be written in the form

$$\left(\frac{d}{dt}+i\Delta+\tfrac{1}{2}\Gamma_{21}\right)\sigma_{12}(t)=\tfrac{1}{2}i\omega_R(t)n(t)\,, \tag{5}$$

$$\left(\frac{d}{dt}+\Gamma_2\right)n(t)=-\Gamma_2-2\,\mathrm{Im}[\omega_R^*(t)\sigma_{12}(t)]\,, \tag{6}$$

where $n=\sigma_{22}-\sigma_{11}$ is the population difference and the normalization condition is $\sigma_{11}+\sigma_{22}=1$. In the above equations, $\Delta=\omega-\omega_{21}$ is the detuning from resonance, Γ_2 the spontaneous lifetime of state $|2\rangle$, and Γ_{21} the width of the resonance which may in general be different from Γ_2 as is the case of elastic collisions. The interaction parameter $\omega_R(t)=2\hbar^{-1}\mu_{12}\epsilon(t)$ is a stochastic process and its root-mean-square value $\overline{\omega}_R=2\hbar^{-1}\mu_{12}\epsilon_0$ will herein be referred to as the average Rabi oscillation frequency. Equations (5) and (6) are stochastic differential equations. What we now need is the average value of their solution. Integrating both of these equations formally and eliminating $\sigma_{12}(t)$, we obtain the integral equation

$$n(t)=-1-\mathrm{Re}\int_0^t e^{\Gamma_2(t_1-t)}dt_1 \times\int_0^{t_1}\exp[i\Delta+\tfrac{1}{2}\Gamma_{21}](t_2-t_1) \times\omega_R^*(t_1)\omega_R(t_2)n(t_2)\,dt_2\,, \tag{7}$$

where we have used the initial condition $\sigma_{11}(0)=1$ and $\sigma_{22}(0)=\sigma_{12}(0)=0$. Next we calculate the stochastic average of Eq. (7) for the two different stochastic fields and we compare the results.

A. Phase-diffusion field

Taking the stochastic average of Eq. (7) with respect to the fluctuating phase, we find

$$\langle n(t)\rangle=-1-\mathrm{Re}\int_0^t e^{\Gamma_2(t_1-t)}dt_1 \times\int_0^{t_1}\exp\{[i\Delta+\tfrac{1}{2}(\Gamma_{21}+\gamma)](t_2-t_1)\} \times\overline{\omega}_R^2\langle n(t_2)\rangle\,dt_2\,. \tag{8}$$

In deriving the above equation we have used the relation

$$\begin{aligned}\langle\omega_R^*(t_1)\omega_R(t_2)n(t_2)\rangle &=\int_{-\infty}^{\infty}d\phi_1\cdots\int_{-\infty}^{\infty}d\phi_n f(\phi_1\cdots\phi_n;t_1\cdots t_n)\overline{\omega}_R^2 \times e^{-i(\phi_1-\phi_2)}n(\phi_2\cdots\phi_n)\\ &=\left(\overline{\omega}_R^2\int_{-\infty}^{\infty}d\phi_1 f(\phi_1,t_1|\phi_2,t_2)e^{-i(\phi_1-\phi_2)}\right)\\ &\quad\times\left(\int_{-\infty}^{\infty}d\phi_2\cdots\int_{-\infty}^{\infty}d\phi_n f(\phi_2\cdots\phi_n;t_2\cdots t_n)\times n(\phi_2\cdots\phi_n)\right)\\ &=\langle\omega_R^*(t_1)\omega_R(t_2)\rangle\langle n(t_2)\rangle\,,\end{aligned} \tag{9}$$

where

$$f(\phi_1\cdots\phi_n;t_1\cdots t_n),\quad t_1>t_2>\cdots>t_n=0,$$

is the joint probability density of the infinite sequence of random variables $\phi_j=\phi(t_j)$ and[18]

$$f(\phi_1,t_1|\phi_2,t_2)=\frac{f(\phi_1\cdots\phi_n;t_1\cdots t_n)}{f(\phi_2\cdots\phi_n;t_2\cdots t_n)}=\frac{\exp[-(\phi_1-\phi_2)^2/2\gamma(t_1-t_2)]}{[2\pi\gamma(t_1-t_2)]^{1/2}} \tag{10}$$

is the conditional probability density of the Markov process $\phi(t)$. As we have pointed out previously,[16] the decorrelation

$$\langle\omega_R^*(t_1)\omega_R(t_2)n(t_2)\rangle=\langle\omega_R^*(t_1)\omega(t_2)\rangle\langle n(t_2)\rangle$$

in the case of the phase-diffusion field is mathematically rigorous and not simply plausible as argued in Ref. 10. The reason for its validity is the statistical independence of the increments of a Wiener-Levy process. We should point out that the phase-diffusion field has been treated rigorously by two other methods, one using the Fokker-Planck formalism for Markov processes[11] and the other the statistical properties of $\dot{\phi}(t)$ which is white Gaussian noise.[8]

Taking the Laplace transform of both sides of Eq. (8) we find

$$\langle N(p)\rangle=-\frac{1}{p}-\mathrm{Re}\frac{\overline{\omega}_R^2}{(p+\Gamma_2)[p+i\Delta+\frac{1}{2}(\Gamma_{21}+\gamma)]}\langle N(p)\rangle\,, \tag{11}$$

where $\langle N(p)\rangle$ is the Laplace transform of $\langle n(t)\rangle$. Using the final value theorem for the Laplace transform [$\lim_{p\to 0}p\langle N(p)\rangle=\langle n(t=\infty)\rangle$], we calculate the steady state value of the population difference

$$\langle n\rangle^{\mathrm{PD}}=-1/(1+S)\,, \tag{12}$$

where

$$S=(\overline{\omega}_R^2/\Gamma_2)\tfrac{1}{2}(\Gamma_{21}+\gamma)/[\Delta^2+\tfrac{1}{4}(\Gamma_{21}+\gamma)^2] \tag{13}$$

is the familiar saturation parameter from the monochromatic theory of a two-level atom. The only change introduced by the fluctuating phase is the addition of the field bandwidth γ to the atomic linewidth Γ_{21}. The resonance curve for the average population of state $|2\rangle$,

$$\langle\sigma_{22}\rangle^{\mathrm{PD}}=\frac{\frac{1}{2}S}{1+S}=\frac{\frac{1}{4}(\Gamma_{21}+\gamma)^2}{\Delta^2/(1+S_0)+\frac{1}{4}(\Gamma_{21}+\gamma)^2}\;\frac{\frac{1}{2}S_0}{1+S_0}\,, \tag{14}$$

where S_0 is the value of S for $\Delta=0$, is Lorentzian and its FWHM is equal to $\sqrt{1+S_0}(\Gamma_{21}+\gamma)$. As we will see in Secs. II B and II C, in the case of a chaotic field this resonance curve is not Lorentzian and its shape depends on the relative magnitude of $\overline{\omega}_R$, Γ_2, Γ_{21}, and γ.

B. Chaotic field

1. Zero bandwidth

a. One-photon resonance. Recall that for phase diffusion, one obtains the simple algebraic relation

$$\langle\omega_R^*(t_1)\omega_R(t_2)n(t_2)\rangle=\langle\omega_R^*(t_1)\omega_R(t_2)\rangle\langle n(t_2)\rangle$$

which enables one to convert the stochastic average of Eq. (7) into an integral equation for $\langle n(t)\rangle$ solvable by Laplace transform. The result is an exact expression in closed form for $\langle n(t)\rangle$. For a chaotic field, the calculation of the stochastic average of Eq. (7) is in general much more difficult. Only in the case of zero bandwidth are we able to obtain a nonperturbative relation between the correlation $\langle\omega_R^*(t_1)\omega_R(t_2)n(t_2)\rangle$ and the average value $\langle n(t_2)\rangle$ of the population difference. To obtain such a relation we consider the formal expression for $\langle n(t)\rangle$. Note that for $\gamma=0$, —which implies infinite correlation time—the field is a random variable with statistics independent of time. The real amplitude $|\epsilon|$ of the field has a Rayleigh distribution while the phase ϕ has a distribution uniform from 0 to 2π. The intensity of the chaotic field has an exponential distribution. Thus the average of $n(t)$ is formally given by

$$\langle n(t)\rangle=\int_0^{2\pi}\int_0^{\infty}\frac{2|\omega_R|e^{-(|\omega_R|/\overline{\omega}_R)^2}}{2\pi\overline{\omega}_R^2}\times n(|\omega_R|,\phi,t)d|\omega_R|\,d\phi\,. \tag{15}$$

If we now take the derivative of both sides of Eq. (15) with respect to $\overline{\omega}_R^2=\langle\omega_R^*\omega_R\rangle$, we obtain

$$\langle\omega_R^*\omega_R n(t)\rangle=\langle\omega_R^*\omega_R\rangle\langle n(t)\rangle+\langle\omega_R^*\omega_R\rangle^2\frac{d\langle n(t)\rangle}{d\langle\omega_R^*\omega_R\rangle}\,. \tag{16}$$

The first term on the right-hand side corresponds to the decorrelation result obtained for the phase-diffusion field. By simply moving this term to the left-hand side, we see that the second term is equal to the correlation $\langle\omega_R^*\omega_R\delta n(t)\rangle$ between the random intensity of the chaotic field ($|\omega_R|^2$) and the stochastic fluctuation $\delta n(t)=n(t)-\langle n(t)\rangle$ of the population difference around its average value $\langle n(t)\rangle$.

If we calculate the average of Eq. (7) using Eq. (16) and then take the Laplace transform, we find that the steady-state value $\langle n(t=\infty)\rangle$ satisfies the differential equation

$$S^2\frac{d\langle n\rangle}{dS}+(1+S)\langle n\rangle+1=0\,, \tag{17}$$

where S is the saturation parameter defined in Eq. (13) with $\gamma=0$. It is worth noting for the sake of comparison that for phase diffusion, the Laplace transform yields an algebraic expression for $\langle n(\infty)\rangle$ while in this case it leads to a differential equation satisfied by $\langle n(\infty)\rangle$. Its solution [with the boundary condition $\langle n(S=0)\rangle=-1$] can be written in the various forms

$$\langle n\rangle^{\mathrm{CH}}=-\frac{e^{1/S}}{S}\int_1^{\infty}\frac{e^{-t/S}}{t}\,dt$$
$$\equiv-\frac{e^{1/S}}{S}E_1(1/S)=\int_0^{\infty}\left(\frac{-1}{1+S'}\right)\frac{e^{-S'/S}}{S}\,dS'\,, \tag{18}$$

where the function $E_1(1/S)$ is the first exponential integral.[20] For $S\ll 1$ we use the asymptotic expansion of $E_1(1/S)$ which gives $\langle n\rangle^{\mathrm{CH}}=-\sum_{k=0}^{\infty}k!(-S)^k$. while for the phase-diffusion model we can write the series expansion $\langle n\rangle^{\mathrm{PD}}=-\sum_{k=0}^{\infty}(-S)^k$ of Eq. (12). As expected, the two results agree only to first order in perturbation theory. Higher-order terms differ by the factors $\langle|\epsilon|^{2k}\rangle^{\mathrm{CH}}/\langle|\epsilon|^{2k}\rangle^{\mathrm{PD}}=k!$. For large values of S, the series expansion[20] of $E_1(1/S)$ gives $\langle n\rangle^{\mathrm{CH}}\simeq-(\ln S)/S$, while Eq. (12) gives $\langle n\rangle^{\mathrm{PD}}\simeq-1/S$. Clearly, the chaotic field is less effective than the coherent field in saturating a one-photon transition.

The last form of the solution in Eq. (18) shows explicitly that for a zero-bandwidth chaotic field, $\langle n\rangle^{\mathrm{CH}}$ can also be calculated by first solving the problem for a pure coherent state (phase-diffusion field with zero bandwidth) or even a photon-number state and then averaging the resulting expression over the exponential intensity distribution of the chaotic field. This result is well known and we could have used it from the beginning, by first doing the time integral in Eq. (7) and then performing the statistics which for $\gamma=0$ are independent of time. However, the method used here in obtaining Eq. (18) is more general, because for $\gamma\neq 0$ the probability densities are time dependent and the stochastic average must be performed before the time integral. In addition, this approach allows us to compare the decorrelation result of Eq. (9) for the phase-diffusion field with the result of Eq. (16) for the chaotic field.

Another important parameter in describing fluctuations is their standard deviation. We calculate here the standard deviation

$$(\langle\sigma_{22}^2\rangle - \langle\sigma_{22}\rangle^2)^{1/2} = \tfrac{1}{2}(\langle n^2\rangle - \langle n\rangle^2)^{1/2}.$$

This relation is evident if one notes that $\sigma_{22} = \frac{1}{2}(n+1)$. The average of n^2 is given by

$$\langle n^2\rangle = \int_0^\infty \frac{1}{(1+S')^2}\frac{e^{-S'/S}}{S}\,dS' = \frac{e^{1/S}}{S}\int_1^\infty \frac{e^{-t/S}}{t^2}\,dt \equiv \frac{e^{1/S}}{S}E_2\left(\frac{1}{S}\right), \quad (19)$$

where we have simply averaged the value of n^2 for a coherent field over the exponential intensity distribution of the chaotic field. The function $E_2(1/S)$ defined above is the second exponential integral.[20] The standard deviation of σ_{22} is

$$(\langle\sigma_{22}^2\rangle - \langle\sigma_{22}\rangle^2)^{1/2} = \frac{1}{2}\left[\frac{e^{1/S}}{S}E_2\left(\frac{1}{S}\right) - \frac{e^{2/S}}{S^2}E_1^2\left(\frac{1}{S}\right)\right]^{1/2}. \quad (20)$$

For $S \ll 1$, the standard deviation increases with increasing S as $\frac{1}{2}S$. It reaches a maximum value of ~0.12 for $S \simeq 2$, and then decreases as $\frac{1}{2}(1/\sqrt{S})(1-\ln^2 S/S)^{1/2}$ for $S \gg 1$. The decrease of the fluctuations with increasing intensity (S) can be understood as follows. The fluctuations are caused by the random intensity of the chaotic field which has an exponential distribution from zero to infinity. As the average value of the intensity increases and the exponential distribution broadens, the statistical weight of the low intensities which do not cause saturation decreases. Thus, as the average intensity increases, the average value of σ_{22} goes to $\frac{1}{2}$, while the fluctuations go to zero.

b. Two-photon resonance. The method used above to study the saturation behavior of a one-photon resonance in a stochastic field can be easily extended to the case of a multiphoton resonance. We examine here briefly the saturation of a two-photon resonance and compare it to that of a one-photon resonance. Neglecting optical Stark shifts, the equations for the density matrix elements in the two-level model for a two-photon resonance are the same as Eqs. (5) and (6), with $\Delta = 2\omega - \omega_{21}$ and $\omega_R(t) = 2\hbar^{-2}r_{12}\epsilon^2(t)$, where r_{12} is the composite matrix element for a two-photon transition.[15] The steady-state value of the population difference for a coherent field is $n = -1/(1+S)$, where S is the saturation parameter [Eq. (13)] for a two-photon transition. Note that now S is proportional to the square of the intensity. Thus the average value of n for a chaotic field can be obtained from

$$\langle n\rangle^{\mathrm{CH}} = \int_0^\infty \frac{-1}{1+S'}\frac{\exp[-(S'/S)^{1/2}]}{\sqrt{S}}\,d\sqrt{S'} = -\frac{1}{\sqrt{S}}\left[\sin\left(\frac{1}{\sqrt{S}}\right)\mathrm{Ci}\left(\frac{1}{\sqrt{S}}\right) - \cos\left(\frac{1}{\sqrt{S}}\right)\mathrm{si}\left(\frac{1}{\sqrt{S}}\right)\right], \quad (21)$$

where $\mathrm{Ci}(1/\sqrt{S})$ and $\mathrm{si}(1/\sqrt{S})$ are the cosine and sine integrals, respectively.[20] For $S \ll 1$ the asymptotic expansions of Eq. (21) gives $\langle n\rangle^{\mathrm{CH}} = -\sum_{k=0}^\infty (2k)!(-S)^k$, while for the phase-diffusion field we have $\langle n\rangle^{\mathrm{PD}} = -\sum_{k=0}^\infty(-S)^k$, as in the case of a one-photon resonance. To first order in S, and thus to second order in the intensity of the field, we have $\langle\sigma_{22}\rangle^{\mathrm{CH}}/\langle\sigma_{22}\rangle^{\mathrm{PD}} = 2!$. This is the well known 2! enhancement of two-photon absorption in a weak chaotic field relative to a coherent field of the same average intensity.[21,22] For large values of S, the series expansion[20] of Eq. (21) gives $\langle n\rangle^{\mathrm{CH}} \simeq -\pi/(2\sqrt{S})$, while for the phase-diffusion field we have $\langle n\rangle^{\mathrm{PD}} \simeq -1/S$. Thus, the chaotic field is less effective than a coherent field in saturating a two-photon transition. Comparing the one- and two-photon resonances, we see that the chaotic field relative to the coherent field is less effective in saturating a two-photon transition than a one-photon transition. In fact, the higher the order of the multiphoton resonance, the less effective (relative to the coherent field) the chaotic field becomes in saturating the transition. It can be easily shown that under strong saturation ($S^{1/N} \gg 1$) of an N-photon resonance ($N > 1$), $\langle n\rangle^{\mathrm{CH}} \simeq -S^{-1/N}(\pi/N)/\sin(\pi/N)$ and $\langle n\rangle^{\mathrm{PD}} \simeq -1/S$, where the saturation parameter S is proportional to the Nth power of the intensity. Note that regardless of the value of N, $\langle n\rangle^{\mathrm{CH}}$ is inversely proportional to the intensity, while $\langle n\rangle^{\mathrm{PD}}$ is inversely proportional to the Nth power of the intensity.

2. *Arbitrary bandwidth*

As mentioned earlier, for a chaotic field with nonzero bandwidth, we cannot find a nonperturbative relation between $\langle\omega_R^*(t_1)\omega_R(t_2)n(t_2)\rangle$ and $\langle n(t_2)\rangle$ by simply knowing the statistics of $\omega_R(t) = 2\hbar^{-1}\mu_{12}\epsilon(t)$. To see the difficulty involved, consider the formal expression

$$\langle\epsilon^*(t_1)\epsilon(t_2)n(t_2)\rangle = \int_{-\infty}^\infty d\epsilon_{x_1}\int_{-\infty}^\infty d\epsilon_{y_1}\cdots\int_{-\infty}^\infty d\epsilon_{x_n}\int_{-\infty}^\infty d\epsilon_{y_n} f(\epsilon_{x_1}\cdots\epsilon_{x_n};t_1\cdots t_n) f(\epsilon_{y_1}\cdots\epsilon_{y_n};t_1\cdots t_n)(\epsilon_{x_1}-i\epsilon_{y_1})(\epsilon_{x_2}+i\epsilon_{y_2})n(\epsilon_{x_1},\epsilon_{y_1}\cdots\epsilon_{x_n},\epsilon_{y_n}), \quad (22)$$

where

$$\epsilon(t)=\epsilon_x(t)+i\epsilon_y(t),$$

and

$$f(\epsilon_{x_1}\cdots\epsilon_{x_n};t_1\cdots t_n),$$

$$f(\epsilon_{y_1}\cdots\epsilon_{y_n};t_1\cdots t_n),\quad t_1>t_2>\cdots>t_n=0,$$

are Gaussian joint probability densities of the infinite sequences of the random variables $\epsilon_{x_j}=\epsilon_x(t_j)$ and $\epsilon_{y_j}=\epsilon_y(t_j)$, respectively. Using the Markov property,

$$f(\epsilon_{x_1}\cdots\epsilon_{x_n};t_1\cdots t)=f(\epsilon_{x_1},t_1|\epsilon_{x_2},t_2)\times f(\epsilon_{x_2}\cdots\epsilon_{x_n};t_2\cdots t_n),$$

where the conditional density is given by[18]

$$f(\epsilon_{x_1},t_1|\epsilon_{x_2},t_2)=\frac{\exp[-(\epsilon_{x_1}-r\epsilon_{x_2})^2/2\langle\epsilon_{x_1}^2\rangle(1-r^2)]}{[2\pi\langle\epsilon_{x_1}^2\rangle(1-r^2)]^{1/2}},$$

with

$$r=\frac{\langle\epsilon_{x_1}\epsilon_{x_2}\rangle}{[\langle\epsilon_{x_1}^2\rangle\langle\epsilon_{x_2}^2\rangle]^{1/2}}=\exp[-\tfrac{1}{2}\gamma(t_1-t_2)]$$

one can prove that

$$\langle\omega_R^*(t_1)\omega_R(t_2)n(t_2)\rangle=\exp[-\tfrac{1}{2}\gamma(t_1-t_2)]\langle\omega_R^*(t_2)\omega_R(t_2)n(t_2)\rangle\,. \tag{23}$$

But no further progress can be made. [Relations similar to Eq. (23) will be used in Sec. III.] We can, however, obtain a perturbation series expansion of the correlation $\langle\omega_R^*(t_1)\omega_R(t_2)n(t_2)\rangle$, for any general stochastic field. As we will see below, for a Markovian chaotic field the series can be summed systematically to all orders.

To develop a summable series it is convenient to write the stochastic population difference as[23]

$$n(t)=\langle n(t)\rangle+\delta n(t) \tag{24}$$

from which it is obvious that $\langle\delta n(t)\rangle=0$. Taking the formal stochastic average of Eq. (7) and using Eq. (24) we write

$$\langle n(t)\rangle=-1-\mathrm{Re}\int_0^t e^{\Gamma_2(t_1-t)}dt_1\int_0^{t_1}\exp[(i\Delta+\tfrac{1}{2}\Gamma_{21})(t_2-t_1)]\times[\langle\omega_R^*(t_1)\omega_R(t_2)\rangle\langle n(t_2)\rangle+\langle\omega_R^*(t_1)\omega_R(t_2)\delta n(t_2)\rangle]\,dt_2\,, \tag{25}$$

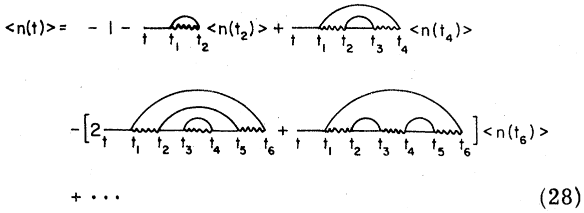
and subtracting Eq. (25) from Eq. (7) we find

$$\delta n(t_2)=-\mathrm{Re}\int_0^{t_2}e^{\Gamma_2(t_3-t_2)}dt_3\int_0^{t_3}\exp[(i\Delta+\tfrac{1}{2}\Gamma_{21})(t_4-t_3)]\times\{[\omega_R^*(t_3)\omega_R(t_4)-\langle\omega_R^*(t_3)\omega_R(t_4)\rangle]\langle n(t_4)\rangle+[\omega_R^*(t_3)\omega_R(t_4)\delta n(t_4)-\langle\omega_R^*(t_3)\omega_R(t_4)\delta n(t_4)\rangle]\}\,dt_4\,. \tag{26}$$

The fluctuation $\delta n(t_2)$ can be calculated in terms of $\langle n(t)\rangle$ at earlier times by iteration. Iterating Eq. (26) and eliminating $\delta n(t_2)$ in Eq. (25) we obtain the series integral equation

$$\begin{aligned}\langle n(t)\rangle=&-1-\mathrm{Re}\int_0^t e^{\Gamma_2(t_1-t)}dt_1\int_0^{t_1}\exp[(i\Delta+\tfrac{1}{2}\Gamma_{21})(t_2-t_1)]\\&\times dt_2\bigg[\langle\omega_R^*(t_1)\omega_R(t_2)\rangle\langle n(t_2)\rangle-\mathrm{Re}\int_0^{t_2}e^{\Gamma_2(t_3-t_2)}dt_3\int_0^{t_3}\exp[(i\Delta+\tfrac{1}{2}\Gamma_{21})(t_4-t_3)]dt_4\\&\times\bigg(\langle\omega_R^*(t_1)\omega_R(t_4)\rangle\langle\omega_R^*(t_3)\omega_R(t_2)\rangle\langle n(t_4)\rangle-\mathrm{Re}\int_0^{t_4}e^{\Gamma_2(t_5-t_4)}dt_5\int_0^{t_5}\exp[(i\Delta+\tfrac{1}{2}\Gamma_{21})(t_6-t_5)]dt_6\\&\times\{[\langle\omega_R^*(t_1)\omega_R(t_6)\rangle\langle\omega_R^*(t_3)\omega_R(t_4)\rangle\langle\omega_R^*(t_5)\omega_R(t_2)\rangle+\langle\omega_R^*(t_1)\omega_R(t_4)\rangle\langle\omega_R^*(t_3)\omega_R(t_6)\rangle\langle\omega_R^*(t_5)\omega_R(t_2)\rangle\\&+\langle\omega_R^*(t_1)\omega_R(t_6)\rangle\langle\omega_R^*(t_3)\omega_R(t_2)\rangle\langle\omega_R^*(t_5)\omega_R(t_4)\rangle]\langle n(t_6)\rangle-\cdots\}\bigg)\bigg]\,.\end{aligned} \tag{27}$$

For a Markovian chaotic field, the first-order correlation function is exponential and the above equation can be solved by Laplace transform. For a nonexponential correlation function, the Laplace transform is not useful because it involves convolutions of the Laplace transforms of the correlation functions and the unknown $\langle n(t)\rangle$. This limits us to Markovian stochastic fields and Lorentzian line shapes. The Laplace transform of Eq. (27) can be calculated in a systematic way by writing that equation in terms of diagrams[13,14,23]



(28)

where the straight and wavy line segments between two successive vertices at t_j and t_{j+1} are associated with the factors $\exp[\Gamma_2(t_{j+1}-t_j)]$ and $\frac{1}{2}\exp[(i\Delta+\frac{1}{2}\Gamma_{21})(t_{j+1}-t_j)]+\text{c.c.}$, respectively. The loop connecting two vertices at t_j and t'_j, where $t_j > t'_j$ ($j<j'$), is associated with the factor $\overline{\omega}_R^2\exp[-\frac{1}{2}\gamma(t_j-t_{j'})]$; the vertex at t_j is called the initial vertex of the loop and the one at t'_j is called the final vertex. As we can see from Eq. (27), a diagram may contain intersecting loops

$$[\langle\omega_R^*(t_1)\omega_R(t_4)\rangle\langle\omega_R^*(t_3)\,\omega_R(t_6)\rangle\langle\omega_R^*(t_5)\omega_R(t_2)\rangle]\,,$$

but because of the exponential form of the first-order correlation function it can be replaced by an equivalent diagram that does not contain intersecting loops

$$[\langle\omega_R^*(t_1)\omega_R(t_6)\rangle\langle\omega_R^*(t_3)\,\omega_R(t_4)\rangle\langle\omega_R^*(t_5)\omega_R(t_2)\rangle]\,.$$

The number of equivalent diagrams of a particular kind is found as follows. To each vertex t_j we associate a number $k_j=k_{j-1}+1$, if it is an initial vertex, and $k_j=k_{j-1}-1$, if it is a final vertex. The first initial vertex at t_1 is given the number $k_1=1(k_0=0)$. The number of equivalent diagrams equals the product of the numbers $\frac{1}{2}(k_j+1)$, if k_j is odd, and $\frac{1}{2}k_j$, if k_j is even, for all initial vertices. Note that only irreducible diagrams appear in Eq. (28). A diagram is irreducible if it cannot be subdivided into lower-order diagrams. If instead of iterating Eq. (26) for $\delta n(t)$ we had iterated Eq. (7) for $n(t)$ as was done in Ref. 13, there would be both reducible and irreducible diagrams but without the factor $\langle n(t_{2m})\rangle$. In Eq. (28) the term corresponding to the first diagram is equivalent to all the reducible diagrams obtained by following the method of Ref. 13.

Taking the Laplace transform of Eq. (28) by repeated application of the frequency-shift theorem we find

$$\langle N(p)\rangle=-(1/p)/[1+\Sigma_1(p)]\,, \tag{29}$$

where $\Sigma_1(p)$ is the first irreducible function corresponding to the set of diagrams in Eq. (28) and is given by the relation

$$\begin{aligned}\Sigma_1(p)&=\text{Re}\,\frac{\overline{\omega}_R^2}{(p+\Gamma_2)[p+i\Delta+\frac{1}{2}(\Gamma_{21}+\gamma)]}\\&\quad\times\left(1-\text{Re}\,\frac{\overline{\omega}_R^2}{(p+\Gamma+\gamma)[p+i\Delta+\frac{1}{2}(\Gamma_{21}+\gamma)]}+\cdots\right)\\&=\text{Re}\,\frac{\overline{\omega}_R^2}{(p+\Gamma_2)[p+i\Delta+\frac{1}{2}(\Gamma_{21}+\gamma)]}\;\frac{1}{1+\Sigma_2(p)}\,,\end{aligned} \tag{30}$$

which defines the second irreducible function $\Sigma_2(p)$. The diagrammatic representation of $\Sigma_2(p)$ consists of the set of irreducible diagrams which are obtained by removing the outer loop in the diagrams for $\Sigma_1(p)$. Higher-order irreducible functions are defined in a similar way. The recursion relation for these irreducible functions is

$$\Sigma_m(p)=S_m(p)\{1/[1+\Sigma_{m+1}(p)]\}\,, \tag{31}$$

where

$$S_m(p)=\text{Re}\,\frac{(m+1)\overline{\omega}_R^2}{2[p+\Gamma_2+\frac{1}{2}(m-1)\gamma][p+i\Delta+\frac{1}{2}(\Gamma_{21}+m\gamma)]}\,, \tag{32a}$$

for m odd, and

$$S_m(p)=\text{Re}\,\frac{m\overline{\omega}_R^2}{2(p+\Gamma_2+\frac{1}{2}m\gamma)\{p+i\Delta+\frac{1}{2}[\Gamma_{21}+(m-1)\gamma]\}} \tag{32b}$$

for m even. The steady-state value of the population difference for a chaotic field of arbitrary bandwidth is given by the continued fraction

$$\langle n\rangle^{\text{CH}}=\frac{-1}{1+\Sigma_1}=\cfrac{-1}{1+\cfrac{S_1}{1+\cfrac{S_2}{1+\cdots}}}\,, \tag{33}$$

where $\Sigma_1=\Sigma_1(p=0)$ is the saturation parameter for a chaotic field and $S_m=S_m(p=0)$ are coefficients in the continued fraction expansion of Σ_1. The first saturation coefficient S_1 is identical to the saturation parameter S [Eq. (13)] for a phase-diffusion field. Since $\Sigma_1<S_1$, a chaotic field is always less effective than a phase-diffusion field in saturating a one-photon transition. For $\gamma=0$, the continued fraction in Eq. (33) is identical to the continued fraction expansion of $(-1/S)e^{1/S}E_1(1/S)$ in Eq. (18). For $\gamma\neq 0$ the continued fraction does not seem to be associated with a familiar function, but it converges and can be suitably truncated and summed to any desired accuracy. The number of coefficients required to reach a certain accuracy increases with the value of S_1.[24]

A similar approach to the problem of a two-level atom in a chaotic field was taken recently in a paper by Przhibelskii,[13] where the equation for $n(t)$ was iterated directly giving both reducible and irreducible diagrams. The contribution of the irreducible diagrams was, however, neglected and as a result the decorrelation

$$\langle \omega_R(t_1)\omega_R(t_2)n(t_2)\rangle = \langle \omega_R^*(t_1)\omega_R(t_2)\rangle\langle n(t_2)\rangle$$

was found to be valid even for a chaotic field. But as we have pointed out previously,[16] and have shown above, the decorrelation is rigorous only for fields whose nth-order correlation function satisfies Eq. (2). Any additional terms on the right-hand side of Eq. (2) will give rise to irreducible diagrams and the decorrelation will no longer be valid. In the case of the phase-diffusion field, Eq. (2) is satisfied because of the independence of the phase increments. Although independence is a sufficient condition for decorrelation, it is not a necessary requirement. Therefore, the decorrelation can be valid for other types of phase fluctuations provided they satisfy Eq. (2). On the other hand, intensity fluctuations can never satisfy Eq. (2), because $\langle I^N\rangle$ is different than $\langle I\rangle^N$. Thus, the decorrelation will never be valid in the presence of intensity fluctuations.

C. Numerical calculations and discussion

To illustrate the differences in the saturation behavior of a two-level atom in a phase-diffusion field from that in a chaotic field with the same average power and spectral width, we have calculated the difference in the population of the excited state for the two fields, $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$. The continued fraction in Eq. (33) was evaluated with twelve hundred coefficients. At the highest saturation ($S_1 = 200$) in these calculations, the absolute error in the value of $\langle\sigma_{22}\rangle^{CH}$ is 2×10^{-5}. For lower values of saturation the error is much smaller. In all of the calculations presented below we have assumed $\Gamma_{21} = \Gamma_2$.

Figure 1 shows the dependence of $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ on the ratio $\bar\omega_R/\Gamma_2$ for different values of the bandwidth γ, under exact resonance. The chaotic field is clearly always less effective than the phase-diffusion field. As $\bar\omega_R/\Gamma_2$ increases, the difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ increases, and after reaching a maximum, decreases. The value and the position of this maximum depend on the bandwidth of the fields. For $\gamma = 0$ the maximum occurs at $\bar\omega_R/\Gamma_2 \simeq 1.4$ and has a value of ~0.068, which corresponds to a 20% difference in the population of the excited state ($\langle\sigma_{22}\rangle^{PD} = 0.398$, $\langle\sigma_{22}\rangle^{CH} = 0.33$). Comparing the curve for $\langle\sigma_{22}\rangle^{CH} - \langle\sigma_{22}\rangle^{PD}$ with the curve for the standard deviation of σ_{22}^{CH} (dashed line), we see that there is a correlation between the two

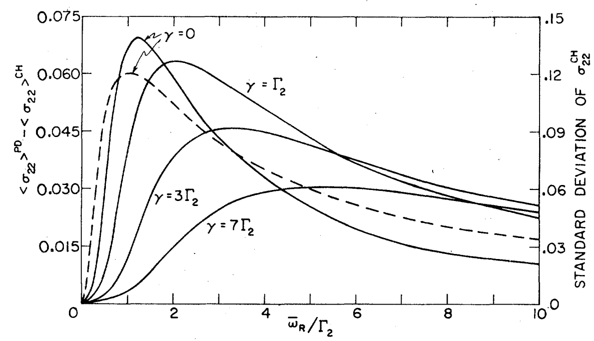


FIG. 1. Plot of the difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ in the population of the excited state for phase diffusion and chaotic fields vs the ratio of the Rabi frequency ($\bar\omega_R$) to the spontaneous decay rate (Γ_2), for different values of the field bandwidth (γ). The dashed line is the standard deviation for σ_{22}^{CH}.

quantities, and that the second is always larger than the first. The standard deviation of σ_{22}^{CH} has a maximum value of ~0.12 at $\bar\omega_R/\Gamma_2 \simeq 1$, where $\langle\sigma_{22}\rangle^{PD} = 0.33$ and $\langle\sigma_{22}\rangle^{CH} = 0.27$. Thus, although on the average the chaotic field is less effective than the phase-diffusion field, there is a large probability that a single measurement of σ_{22}^{CH} will yield a value larger than $\langle\sigma_{22}\rangle^{PD}$ (for $\gamma = 0$ the standard deviation of σ_{22}^{PD} is zero). As the bandwidth is increased from zero and the fluctuations in the fields become more rapid, the position of the maximum difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ shifts to higher values of $\bar\omega_R/\Gamma_2$ and its value decreases. The reason for the shift is that both fields become less effective as γ is increased. The decrease in the value of the maximum difference is caused by the reduced correlation between the atomic and the intensity fluctuations, as the latter become more rapid. Generally, $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ is small when either $S_1 \ll 1$ and first-order perturbation theory holds, or when saturation is very strong and $\langle\sigma_{22}\rangle^{CH} \simeq \langle\sigma_{22}\rangle^{PD} \simeq \frac{1}{2}$.

Figure 2 shows the dependence of $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$

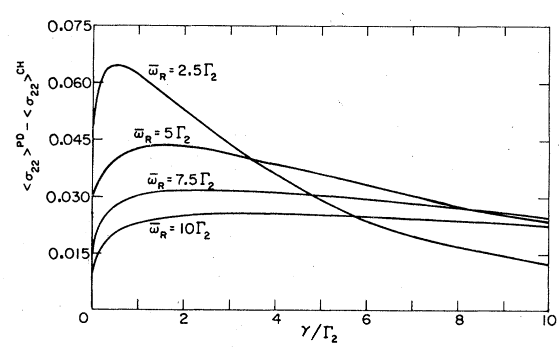


FIG. 2. Plot of $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ vs the ratio of the field bandwidth (γ) to the spontaneous decay rate (Γ_2), for different values of the Rabi frequency ($\bar\omega_R$).

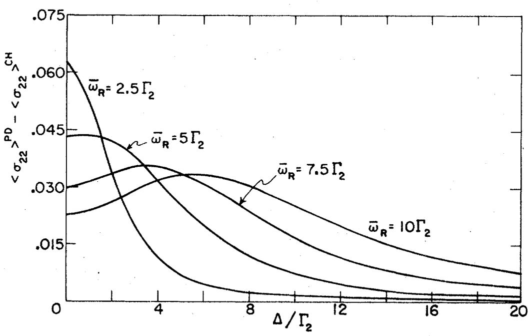


FIG. 3. Plot of $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ vs the ratio of the detuning (Δ) to the spontaneous decay rate (Γ_2), for different values of the Rabi frequency ($\bar\omega_R$).

on the ratio γ/Γ_2 for different values of $\bar\omega_R$; under exact resonance. For $\gamma = 0$, the difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ decreases as $\bar\omega_R$ increases from $2.5\Gamma_2$ to $10\Gamma_2$. As γ is increased, first the difference increases—because the saturation decreases—but then it decreases, because the correlation between the atomic and intensity fluctuations decreases. Figure 3 shows the dependence of $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ on the detuning from resonance for different values of $\bar\omega_R$ with $\gamma = \Gamma_2$. As the detuning increases, the saturation decreases from its value on resonance. If the saturation on resonance is weak, the fluctuations in σ_{22}^{CH} decrease monotonically with increasing Δ, and so does the difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$. If, however, the saturation on resonance is strong, then as the detuning increases and the saturation decreases, the fluctuations in σ_{22}^{CH} and the difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ first increase to a maximum and then decrease. The value of maximum difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ and its position depend on the value of $\bar\omega_R$, Γ_2, and γ. Unlike the resonance curve for $\langle\sigma_{22}\rangle^{PD}$, the resonance curve for $\langle\sigma_{22}\rangle^{CH}$ is not Lorentzian and its shape depends on the value of $\bar\omega_R$, Γ_2, and γ.

The saturation behavior of a one-photon transition can be compared to that of a two-photon transition. Figure 4 shows the dependence of $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ and $\langle\sigma_{22}\rangle^{CH}/\langle\sigma_{22}\rangle^{PD}$ on the saturation parameter for a two-photon transition, in the case of $\gamma = 0$. For $S < \frac{1}{4}$ the chaotic field is seen to be more effective than the phase-diffusion field, while for $S > \frac{1}{4}$ it is the other way around. The maximum difference $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ occurs at $S \simeq 6$ and has a value of ~0.12 ($\langle\sigma_{22}\rangle^{PD} = 0.43$, $\langle\sigma_{22}\rangle^{CH} = 0.31$), almost twice as large as in the case of a one-photon resonance (see Fig. 1). In the weak field limit $S \to 0$, we have $\langle\sigma_{22}\rangle^{CH}/\langle\sigma_{22}\rangle^{PD} = 2$, the well known 2! enhancement of two-photon absorption in a weak chaotic field.[21] As the saturation increases, however, this enhancement decreases. At $S \simeq \frac{1}{4}$, the upper-state populations became equal, $\langle\sigma_{22}\rangle^{CH} = \langle\sigma_{22}\rangle^{PD} = 0.1$. With

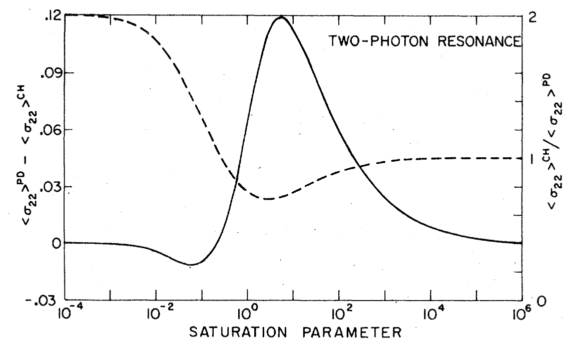


FIG. 4. Plot of $\langle\sigma_{22}\rangle^{PD} - \langle\sigma_{22}\rangle^{CH}$ (solid line) and $\langle\sigma_{22}\rangle^{CH}/\langle\sigma_{22}\rangle^{PD}$ (dashed line) for a two-photon resonance vs the saturation parameter. The bandwidth of the fields is zero.

further increase in S, the ratio $\langle\sigma_{22}\rangle^{CH}/\langle\sigma_{22}\rangle^{PD}$ continues to decrease until it reaches a minimum value of ~0.7 at $S \simeq 3$, and then goes to unity in the limit of $S \to \infty$. For $\gamma \neq 0$, we expect a similar behavior. In the limit of $S \to 0$, we have[22]

$$\frac{\langle\sigma_{22}\rangle^{CH}}{\langle\sigma_{22}\rangle^{PD}} = 2\,\frac{\Gamma_{21} + 2\gamma}{\Gamma_{21} + 4\gamma}\,\frac{\Delta^2 + \frac{1}{4}(\Gamma_{21} + 4\gamma)^2}{\Delta^2 + \frac{1}{4}(\Gamma_{21} + 2\gamma)^2}\,,$$

which for $\Delta = 0$ and $\gamma \gg \Gamma_{21}$ has a maximum value of 4. With increasing saturation the ratio $\langle\sigma_{22}\rangle^{CH}/\langle\sigma_{22}\rangle^{PD}$ will decrease to a minimum below unity and then will tend to unity, as in the case of $\gamma = 0$.

III. STARK SPLITTING IN DOUBLE RESONANCE

We consider a three-level atom, with levels labeled $|1\rangle$, $|2\rangle$, and $|3\rangle$, and respective energies $\hbar\omega_1$, $\hbar\omega_2$, and $\hbar\omega_3 (\omega_1 < \omega_2 < \omega_3)$. The transitions $|1\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ are assumed to have electric dipole moments μ_{12} and μ_{23}, respectively, while the transition $|1\rangle \leftrightarrow |3\rangle$ is dipole forbidden. The atom is interacting with two stochastic fields $E_a(t) = \epsilon_a(t)e^{i\omega_a t} + \text{c.c.}$ and $E_b(t) = \epsilon_b(t)e^{i\omega_b t} + \text{c.c.}$, whose center frequencies ω_a and ω_b are (near) resonant with the transitions $|1\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |3\rangle$, respectively. In a recent paper,[16] we studied the case in which $E_a(t)$ and $E_b(t)$ are two uncorrelated phase-diffusion fields. In this section, we study the case in which $E_a(t)$ and $E_b(t)$ are two uncorrelated Markovian chaotic fields, with spectral widths γ_a and γ_b, respectively.

The relevant equations for the density matrix elements of a three-level system in the rotating wave approximations [i.e., $\rho_{12}(t) = \sigma_{12}(t)e^{i\omega_a t}$, $\rho_{23}(t) = \sigma_{23}(t)e^{i\omega_b t}$, $\rho_{13}(t) = \sigma_{13}(t)e^{i(\omega_a + \omega_b)t}$, $\rho_{ii}(t) = \sigma_{ii}(t)$, where the $\sigma_{ij}(t)$ are slowly varying amplitudes] are given in Ref. 16. The steady-state value of the population of level $|3\rangle$, averaged over the fluctuations of both fields is given by

$$\langle\langle\sigma_{33}\rangle_b\rangle_a = (1/\Gamma_3)\,\mathrm{Re}(i\langle\langle\omega^*_{\mathrm{Rb}}\sigma_{23}\rangle_b\rangle_a)\,, \tag{34}$$

where Γ_3 is the lifetime of level $|3\rangle$, and $\omega_{\mathrm{Rb}}(t) = 2\hbar^{-1}\mu_{23}\epsilon_b(t)$ is the interaction parameter for the $|2\rangle \leftrightarrow |3\rangle$ transition. The stochastic average $\langle\langle\omega^*_{\mathrm{Rb}}(t)\sigma_{23}(t)\rangle_b\rangle_a$ can be calculated from the stochastic integral equation[16]

$$\begin{aligned}\omega^*_{\mathrm{Rb}}(t)\sigma_{23}(t) &= -\frac{i}{2}\int_0^t \exp[(i\Delta_b+\tfrac{1}{2}\Gamma_{32})(t_1-t)]\,\omega^*_{\mathrm{Rb}}(t)\omega_{\mathrm{Rb}}(t_1)[\sigma_{22}(t_1)-\sigma_{33}(t_1)]\,dt_1\\ &\quad+\frac{1}{4}\int_0^t \exp[(i\Delta_b+\tfrac{1}{2}\Gamma_{32})(t_1-t)]\,dt_1\int_0^{t_1}\exp\{[i(\Delta_a+\Delta_b)+\tfrac{1}{2}\Gamma_{31}](t_2-t_1)\}\\ &\qquad\times[\omega^*_{\mathrm{Rb}}(t)\omega_{\mathrm{Rb}}(t_2)\omega^*_{\mathrm{Ra}}(t_1)\sigma_{12}(t_2)-\omega^*_{\mathrm{Ra}}(t_1)\omega_{\mathrm{Ra}}(t_2)\omega^*_{\mathrm{Rb}}(t)\sigma_{23}(t_2)]\,dt_2\,,\end{aligned} \tag{35}$$

where $\Delta_a = \omega_{21} - \omega_a$ and $\Delta_b = \omega_{32} - \omega_b$ are the detunings from resonance, Γ_{32} and Γ_{31} the relaxation rates of σ_{23} and σ_{13}, respectively, and $\omega_{\mathrm{Ra}}(t) = 2\hbar^{-1}\mu_{12}\epsilon_a(t)$ the interaction parameter for the $|1\rangle \leftrightarrow |2\rangle$ transition. If the ac Stark splitting of this transition is probed with a field $E_b(t)$ sufficiently weak, we can assume $\overline{\omega}^2_{\mathrm{Rb}} \ll \overline{\omega}^2_{\mathrm{Ra}}, (\Gamma_{21}+\gamma_a)^2, (\Gamma_{32}+\gamma_b)^2, (\Gamma_{31}+\gamma_a+\gamma_b)^2$. We can also assume that the population of state $|3\rangle$ is negligible ($\sigma_{33} \ll \sigma_{11}+\sigma_{22} \simeq 1$) and that the probe field $E_b(t)$ has no effect on the $|1\rangle \leftrightarrow |2\rangle$ transition. The above assumptions constitute the usual weak probe approximation in double resonance. Introducing these assumptions and taking the stochastic average of Eq. (35) over both fields we obtain

$$\begin{aligned}\langle\langle\omega^*_{\mathrm{Rb}}(t)\sigma_{23}(t)\rangle_b\rangle_a &= -\frac{i}{2}\int_0^t \exp\{[i\Delta_b+\tfrac{1}{2}(\Gamma_{32}+\gamma_b)](t_1-t)\}\,\overline{\omega}^2_{\mathrm{Rb}}\langle\sigma_{22}(t_1)\rangle_a\,dt_1\\ &\quad+\frac{1}{4}\int_0^t \exp\{[i\Delta_b+\tfrac{1}{2}(\Gamma_{32}+\gamma_b)](t_1-t)\}\,dt_1\\ &\quad\times\int_0^{t_1}\exp\{[i(\Delta_a+\Delta_b)+\tfrac{1}{2}(\Gamma_{31}+\gamma_a+\gamma_b)](t_2-t_1)\}\;\overline{\omega}^2_{\mathrm{Rb}}\;\langle\omega^*_{\mathrm{Ra}}(t_2)\sigma_{12}(t_2)\rangle_a\,dt_2\\ &\quad-\frac{1}{4}\int_0^t \exp\{[i\Delta_b+\tfrac{1}{2}(\Gamma_{32}+\gamma_b)](t_1-t)\}\,dt_1\int_0^{t_1}\exp\{[i(\Delta_a+\Delta_b)+\tfrac{1}{2}(\Gamma_{31}+\gamma_b)](t_2-t_1)\}\\ &\qquad\times[\langle\omega^*_{\mathrm{Ra}}(t_1)\omega_{\mathrm{Ra}}(t_2)\rangle_a\langle\langle\omega^*_{\mathrm{Rb}}(t_2)\sigma_{23}(t_2)\rangle_b\rangle_a+\langle\omega^*_{\mathrm{Ra}}(t_1)\omega_{\mathrm{Ra}}(t_2)\delta\langle\omega^*_{\mathrm{Rb}}(t_2)\sigma_{23}(t_2)\rangle_b\rangle_a]\,dt_2\,,\end{aligned} \tag{36}$$

where

$$\begin{aligned}\delta\langle\omega^*_{\mathrm{Rb}}(t_2)\sigma_{23}(t_2)\rangle_b &\equiv \langle\omega^*_{\mathrm{Rb}}(t_2)\sigma_{23}(t_2)\rangle_b - \langle\langle\omega^*_{\mathrm{Rb}}(t_2)\sigma_{23}(t_2)\rangle_b\rangle_a\\ &= -\frac{i}{2}\int_0^{t_2}\exp\{[i\Delta_b+\tfrac{1}{2}(\Gamma_{32}+\gamma_b)](t_3-t_2)\}\,\overline{\omega}^2_{\mathrm{Rb}}[\sigma_{22}(t_3)-\langle\sigma_{22}(t_3)\rangle_a]\,dt_3\\ &\quad+\frac{1}{4}\int_0^{t_2}\exp\{[i\Delta_b+\tfrac{1}{2}(\Gamma_{32}+\gamma_b)](t_3-t_2)\}\,dt_3\int_0^{t_3}\exp\{[i(\Delta_a+\Delta_b)+\tfrac{1}{2}(\Gamma_{31}+\gamma_b)](t_4-t_3)\}\\ &\qquad\times\big(\overline{\omega}^2_{\mathrm{Rb}}[\omega^*_{\mathrm{Ra}}(t_3)\sigma_{12}(t_4)-\langle\omega^*_{\mathrm{Ra}}(t_3)\sigma_{12}(t_4)\rangle_a]\\ &\qquad-\tfrac{1}{4}[\omega^*_{\mathrm{Ra}}(t_3)\omega_{\mathrm{Ra}}(t_4)-\langle\omega^*_{\mathrm{Ra}}(t_3)\omega_{\mathrm{Ra}}(t_4)\rangle]\langle\langle\omega^*_{\mathrm{Rb}}(t_4)\sigma_{23}(t_4)\rangle_b\rangle_a\\ &\qquad-\tfrac{1}{4}\{\omega^*_{\mathrm{Ra}}(t_3)\omega_{\mathrm{Ra}}(t_4)\delta\langle\omega^*_{\mathrm{Rb}}(t_4)\sigma_{23}(t_4)\rangle_b-\langle\omega^*_{\mathrm{Ra}}(t_3)\omega_{\mathrm{Ra}}(t_4)[\delta\langle\omega^*_{\mathrm{Rb}}(t_4)\sigma_{23}(t_4)\rangle_b]\rangle_a\}\big)\,dt_4\,.\end{aligned} \tag{37}$$

In obtaining the last two equations from Eq. (35) we have used the relations

$$\langle\omega^*_{\mathrm{Ra}}(t_1)\sigma_{12}(t_2)\rangle_a = \exp[-\tfrac{1}{2}\gamma_a(t_1-t_2)]\langle\omega^*_{\mathrm{Ra}}(t_2)\sigma_{12}(t_2)\rangle_a$$

and

$$\langle\omega^*_{\mathrm{Rb}}(t)\sigma_{23}(t_2)\rangle_b = \exp[-\tfrac{1}{2}\gamma_b(t-t_2)]\langle\omega^*_{\mathrm{Rb}}(t_2)\sigma_{23}(t_2)\rangle$$

which, as pointed out in Sec. II [Eq. (23)], are exact for a Markovian chaotic field. Equation (36) can be solved by iterating Eq. (37) and then eliminating $\delta\langle\omega^*_{Rb}(t_2)\sigma_{23}(t_2)\rangle_b$. Generally the series integral equation obtained by this iteration is very complicated and difficult to sum. However, for strong saturation of the $|1\rangle \rightarrow |2\rangle$ transition, we can neglect the two terms on the right-hand side of Eq. (37) which involve the fluctuations $\sigma_{22}(t) - \langle\sigma_{22}(t)\rangle_a$ and $\omega^*_{Ra}(t)\sigma_{12}(t) - \langle\omega^*_{Ra}(t)\sigma_{12}(t)\rangle_a$. Recall that as discussed in Sec. II, under strong saturation these fluctuations vanish. In that case, Eqs. (36) and (37) take the same form as Eqs. (25) and (26), respectively; the first two terms on the right-hand side of Eq. (36) correspond to the term -1 in Eq. (25).

Applying the diagrammatic technique described in Sec. II, and using the relation

$$\langle\omega^*_{Ra}\sigma_{12}\rangle_a = -\Gamma_2\left(\frac{2\Delta_a}{\Gamma_{21}+\gamma_a} + i\right)\langle\sigma_{22}\rangle_a\,, \tag{38}$$

we find that the steady-state value of the stochastic average of the population of level $|3\rangle$ is given by

$$\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm CH} = \langle\sigma_{22}\rangle_a^{\rm CH}\left(\frac{\Gamma_2 R^*/(R+R^*)+T}{ST}\,\cfrac{1}{1+\cfrac{e_1}{1+\cfrac{e_2}{1+\cdots}}} + \text{c.c.}\right)\frac{\overline{\omega}^2_{Rb}}{4\Gamma_3}\,, \tag{39}$$

where

$$R = i\Delta_a + \tfrac{1}{2}(\Gamma_{21}+\gamma_a)\,,$$

$$S = i\Delta_b + \tfrac{1}{2}(\Gamma_{32}+\gamma_b)\,,$$

$$T = i(\Delta_a+\Delta_b) + \tfrac{1}{2}(\Gamma_{31}+\gamma_a+\gamma_b)\,.$$

The coefficients of the continued fraction are given by

$$e_m = \frac{(m+1)\overline{\omega}^2_{Ra}}{8\{i\Delta_b + \frac{1}{2}[\Gamma_{32}+\gamma_b+(m-1)\gamma_a]\}\{i(\Delta_a+\Delta_b)+\frac{1}{2}[\Gamma_{31}+\gamma_b+m\gamma_a]\}} \tag{40}$$

for m odd, and

$$e_m = \frac{m\overline{\omega}^2_{Ra}}{8\{i\Delta_b + \frac{1}{2}[\Gamma_{32}+\gamma_b+m\gamma_a]\}\{i(\Delta_a+\Delta_b)+\frac{1}{2}[\Gamma_{31}+\gamma_b+(m-1)\gamma_a]\}} \tag{41}$$

for m even. Note that had we not neglected the fluctuations $\sigma_{22}(t) - \langle\sigma_{22}(t)\rangle_a$ and $\omega^*_{Ra}(t)\sigma_{12}(t) - \langle\omega^*_{Ra}(t)\sigma_{12}(t)\rangle_a$ in Eq. (37), there would be additional terms in Eq. (39) also involving continued fractions. In the case of strong saturation, however, the contribution of these other terms is negligible. Equation (39) should be compared with Eq. (44) of our previous paper[16] for the case of phase-diffusion fields, which we rewrite here in the form

$$\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm PD} = \langle\sigma_{22}\rangle_a^{\rm PD}\left(\frac{\Gamma_2 R^*/(R+R^*)+T}{ST+\frac{1}{4}\overline{\omega}^2_{Ra}} + \text{c.c.}\right)\frac{\overline{\omega}^2_{Rb}}{4\Gamma_3} \tag{42}$$

Note that the expressions in parentheses in Eqs. (39) and (42) which determine the shape of the resonance curve for $\langle\langle\sigma_{33}\rangle_b\rangle_a$ as a function of Δ_b are different. In the limit of strong saturation, $\overline{\omega}^2_{Ra} \gg (\Gamma_{32}+\gamma_b)(\Gamma_{31}+\gamma_a+\gamma_b)$, the resonance curve for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm PD}$ has two peaks at the roots of the equation

$$\text{Re}[ST+\tfrac{1}{4}\overline{\omega}^2_{Ra}] \simeq -\Delta_b(\Delta_a+\Delta_b)+\tfrac{1}{4}\overline{\omega}^2_{Ra} = 0\,. \tag{43}$$

The two peaks are separated by $(\Delta_a^2+\overline{\omega}^2_{Ra})^{1/2}$, and their widths are determined by the atomic linewidths and the bandwidth of the fields. In the same limit, the resonance curve for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm CH}$ has two peaks at the roots of the equation

$$\text{Re}\left(ST + \cfrac{\frac{1}{4}\overline{\omega}^2_{Ra}}{1+\cfrac{e_2}{1+\cfrac{e_3}{1+\cdots}}}\right) = 0 \tag{44}$$

Because of the complicated dependence on Δ_b, one cannot find an analytical expression for the roots of Eq. (44), as we are able to do for the roots of Eq. (43). However, from comparing these two equations we expect, in the case of strong saturation, the Stark splitting for a chaotic field to be less than for a phase-diffusion field and our numerical calculations show that this is indeed the case.

The continued fraction in Eq. (39) is very complicated and does not seem possible to express it in terms of known functions. It can however be simplified somewhat if $\gamma_a = 0$, in which case it can be expressed in terms of the exponential integral.[20] In order to explore some of the qualitative features of the process, let us consider that case for which Eq. (39) reduces to

$$\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm CH} = \langle\sigma_{22}\rangle_a^{\rm CH}[(\Gamma_2 R^*/(R+R^*)+T) \times (e^z/\overline{\omega}^2_{Ra})E_1(z) + \text{c.c.}]\overline{\omega}^2_{Rb}/\Gamma_3\,, \tag{45}$$

where $z = 4ST/\overline{\omega}_{Ra}^2$. Moreover, for exact resonance in the $|1\rangle \rightarrow |2\rangle$ transition ($\Delta_a = 0$) and under the conditions

$$\Delta_b^2 \gg (\Gamma_{32}+\gamma_b)(\Gamma_{31}+\gamma_b),$$

$$\Delta_b(\Gamma_{31}+\Gamma_{32}+2\gamma_b)/\overline{\omega}_{Ra}^2 \rightarrow 0,$$

Eq. (45) reduces to

$$\langle\langle\sigma_{33}\rangle_b\rangle_b^{CH} = \langle\sigma_{22}\rangle_a^{CH} 2\pi(|\Delta_b|/\overline{\omega}_{Ra}^2)\exp(-4\Delta_b^2/\overline{\omega}_{Ra}^2)\overline{\omega}_{Rb}^2/\Gamma_3 . \quad (46)$$

This equation should also be a good approximation in the case of $\gamma_a \neq 0$, provided that $\gamma_a/\overline{\omega}_{Ra} \ll 1$. Thus, under the above conditions, the Stark split resonance curve for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$ reproduces the Rayleigh distribution for the amplitude of the chaotic field.[4,6] The Stark splitting predicted by Eq. (46) is equal to $\overline{\omega}_{Ra}/\sqrt{2}$. Generally, in the case of amplitude fluctuations and under the conditions for the validity of Eq. (46), the resonance curve for $\langle\langle\sigma_{33}\rangle_b\rangle_a$ will always reproduce the amplitude distribution. Since the root-mean-square (rms) value of the amplitude is generally different from the most probable value, the magnitude of the Stark splitting will be different from the average Rabi frequency $\overline{\omega}_{Ra}$. For a real Gaussian field with zero mean value, there is no Stark splitting.

We should mention here that Elyutin[14] has recently considered Stark splitting for the case in which $E_a(t)$ is a weak monochromatic field and $E_b(t)$ is a strong chaotic field. This author neglects the relaxation of the excited states and assumes that $\sigma_{11}(t) \simeq 1$ and $\sigma_{22}(t) \simeq \sigma_{23}(t) \simeq 0$. The Stark effect in this case takes place between the two excited states which have no population. For $\Delta_b = 0$, these assumptions lead to a simple stochastic expression for the absorption of the weak monochromatic field, which is then averaged over the fluctuations of the chaotic field. Note that in this case the problem of atom-field correlations does not arise.

Returning now to the general case of arbitrary bandwidth (γ_a), we present numerical calculations in which the predictions of Eqs. (39) and (42) are compared. Figure 5 shows the dependence of the steady-state value of $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$ and $\langle\langle\sigma_{33}\rangle_b\rangle_a^{PD}$ on $\Delta_b/\overline{\omega}_{Ra}$ for two different values of $\overline{\omega}_{Ra}$, in the case of exact resonance ($\Delta_a = 0$). The values of the parameters used in the calculations are shown in Fig. 5, and correspond to the parameters in the recent doubly resonant ($3S_{1/2} \rightarrow 3P_{1/2} \rightarrow 4D_{3/2}$) three-photon ionization experiments in sodium.[17] For the 700-nsec laser pulses used in those experiments, the probability of ionization was less than 10^{-3}. Thus, the approximation $\sigma_{11}(t) + \sigma_{22}(t) + \sigma_{33}(t) = 1$ is justified. The ion signal is approximately proportional to the steady-state value of $\langle\langle\sigma_{33}\rangle_b\rangle_a$. As we can see, the Stark splitting caused by a strong chaotic field is less than that caused by a phase-diffusion field with the same average power. For $\overline{\omega}_{Ra} = 10\gamma_a$, the Stark splittings for the phase-diffusion and the chaotic field are $\overline{\omega}_{Ra}$ and $0.8\overline{\omega}_{Ra}$, respectively. For higher values of $\overline{\omega}_{Ra}$ the Stark splitting for the chaotic field tends to the limit $\overline{\omega}_{Ra}/\sqrt{2}$. The on resonance dip for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$ is very shallow compared to that for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{PD}$. For $\overline{\omega}_{Ra} = 5\gamma_a$, the peak to dip ratio for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{PD}$ is 4.6, while for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$ is only 1.2. For $\overline{\omega}_{Ra} = 10\gamma_a$, these ratios become 17 and 1.63, respectively. The shallow dip and the broadening of the peaks in the case of $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$ are caused by the intensity fluctuations in the chaotic field. If we compare Fig. 5 with Fig. 1(a) of Ref. 17, we find that the experimental data (Stark splitting 8 GHz) is modeled better by $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$ than by $\langle\langle\sigma_{33}\rangle_b\rangle_a^{PD}$, with $\overline{\omega}_{Ra} = 10\gamma_a$. The value of the peak to dip ratio for the experimental curve is equal to 1.5, which is very close to the value of 1.63 for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$. The experimental curve has a FWHM of 17.6 GHz, while the curve for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{CH}$ has a FWHM of 14.1 GHz. The small difference in the values for the peak-to-dip ratio and the FWHM could be explained by taking into account the hyperfine splitting (1.77 GHz) of the $3S_{1/2}$ ground state of sodium, shot-to-shot variations in laser power and the fact that the laser pulses were not square. The only disagreement between theory and experiment is on the absolute value of the average laser intensity. Using the measured value for the Stark splitting (8 GHz) we calculate (chaotic field) the intensity to be 40 KW/cm^2, whereas the measured intensity was ~2 MW/cm^2, a factor of 50 larger. Direct measurements of the absolute intensity are known to be very difficult and there

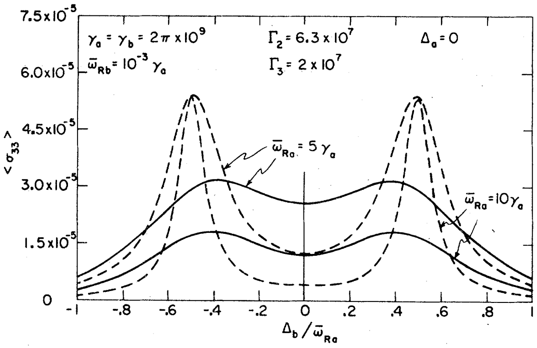


FIG. 5. Plot of the average population $\langle\sigma_{33}\rangle$ for chaotic fields (solid line) and phase-diffusion fields (dashed line) vs the detuning (Δ_b) of the probe field, for two different values of the Rabi frequency $\overline{\omega}_{Ra}$. The strong field is exactly on resonance ($\Delta_a = 0$).

were certainly uncertainties in the measurement, but a factor of 50 seems too large. On the other hand, the actual laser fields were neither ideal chaotic nor Markovian. Most likely the laser of the experiment had a multimode structure. As long as the modes are statistically independent and their separation is sufficiently smaller than the Stark splitting, the discreteness of the modes is not expected to play a role and cannot explain the above discrepancy. The most unfavorable case would have been two modes only correlated in some fashion. But the crudest and most conservative calculation shows that one cannot justify more than a factor of $\sqrt{2}$ discrepancy in that case. Another possibility is that the actual amplitude distribution was such that the most probable value of the field was smaller than the rms value by more than the $\sqrt{2}$ factor for the Rayleigh distribution. This would be consistent with the fact that the FWHM of the experimental curve is larger than that of $\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm CH}$. Such an effective amplitude distribution is generated when one takes into account shot-to shot variations in the laser power. Computer simulation of this problem shows that the intensities below the average value cause the Stark splitting to decrease, while the intensities above the average value cause the FWHM to increase. Thus, when shot-to-shot fluctuations in the laser intensity are taken into account, their average value would have to be higher than 40 kW/cm^2 in order for the splitting to be 8 GHz. This represents an effect in the right direction. But even so, a satisfactory explanation of the above discrepancy must await more detailed experimental data since the necessary detailed information about the intensity, and the model and spectral properties of the laser is not available in this case.

Figure 6 shows the dependence of $\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm CH}$ and $\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm PD}$ on $\Delta_b/\overline{\omega}_{\rm Ra}$ for $\Delta_a=\gamma_a$. As in the case of $\Delta_a=0$, for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm CH}$ the dip between the two peaks is very shallow and the Stark splitting is less than for $\langle\langle\sigma_{33}\rangle_b\rangle_a^{\rm PD}$. The resonance curves for both fields are asymmetric. The asymmetry ratio (peak-to-peak) for a chaotic field is larger than for a phase-diffusion field. For both fields, the asymmetry in the case of $\gamma_a > \Gamma_2$ is opposite from that in the case of $\gamma_a < \Gamma_2$, independently of the value of Δ_a. However, as we have pointed out before,[16] this reversal of the asymmetry depends on the line shape of the field. A field whose line shape falls off faster than a Lorentzian will appear monochromatic (but not necessarily coherent) to the atom beyond a certain detuning, even though $\gamma_a > \Gamma_2$. The asymmetry then becomes the same as for $\gamma_a < \Gamma_2$. This effect was observed in recent experiments[17] with multimode laser pulses whose spectrum was not Lorentzian. Because of the difficulties in treating non-Lorentzian line shapes, we are not able to make a comparison between theory and experiment for $\Delta_a \neq 0$.

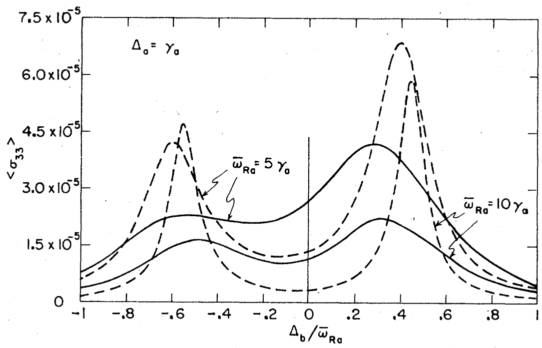


FIG. 6. Same as Fig. 5 but the strong field detuned off resonance ($\Delta_a=\gamma_a$).

IV. CONCLUSIONS

In this paper, we have shown that the presence of amplitude fluctuations affects rather drastically the saturation and Stark splitting of a resonant transition. A surprising new result is that a chaotic field is less effective than a phase-diffusion field in saturating a bound-bound multiphoton transition. It has been known for a long time that in the weak field case, N-photon absorption from a chaotic field of zero bandwidth is larger by a factor of $N!$ than that from a coherent field with the same average power.[21,22] This stems from the fact that below saturation the average N-photon absorption depends only on the Nth-order field correlation and the probability for N-photon coincidence in a chaotic field is $N!$ times that in coherent field. For a Markovian chaotic field, as compared to a phase-diffusion field with the same average power and bandwidth, there is an additional enhancement because the coherence time for the Nth-order field correlation of the chaotic field $[\langle\epsilon^{*N}(t_1)\epsilon^N(t_2)\rangle^{\rm CH} = N!\,\epsilon_0^{2N} e^{-N/2\gamma|t_1-t_2|}]$ is N times larger than that of a phase-diffusion field $(\langle\epsilon^{*N}(t_1)\epsilon^N(t_2)\rangle = \epsilon_0^{2N} e^{-N^2/2\gamma|t_1-t_2|})$.[22] In the saturation regime, however, the average N-photon absorption depends on the infinite sequence of field correlations of order $N^k, k=1,2,\ldots$. It is through this infinite sequence of correlation functions that the atom gains more detailed information about the statistics of the intensity fluctuations in the chaotic field. We must emphasize again that in weak N-photon absorption below saturation, the atom responds only to the Nth-order field correlation, that is one particular average of the field fluctuations. Thus it is only in the saturation regime that the atom can always distinguish between fields with different stochastic properties. A physical interpretation for the chaotic field being less effective than the phase-diffusion field in saturating an atomic transition now seems to emerge. The atom sees the intensity fluctuations of the chaotic field and relaxes between spikes in the intensity, going partially out of saturation. Thus, the same intensity fluctuations which make the chaotic field more effective in the case of weak excitation, make it also less effective in the case of saturation. These results which we have obtained for the chaotic field should also hold to varying degree for other fields with intensity fluctuations.

The effects of intensity fluctuations on the resonant excitation of an atomic transition are demonstrated even more clearly on the Stark splitting of the transition, as observed by weak probing in double resonance. In a one-photon transition the splitting is proportional to the real amplitude of the field. If the amplitude (intensity) of the field fluctuates, the doublet structure is broadened and at low intensities can be washed out completely. For very strong intensities, when the atomic and field linewidths become negligible compared to the average Rabi frequency, the doublet structure reproduces the probability distribution function of the field amplitude. In that case the splitting is determined by the most probable value of the field amplitude and not by its rms value. Thus, the Stark splitting of a transition in a fluctuating field can be used to actually measure the probability distribution function of the field amplitude. Generally, in the case of an N-photon transition the Stark split resonance for very strong fields should reproduce the distribution function for the Nth power of the field amplitude.

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