ac Stark splitting in intense stochastic driving fields with Gaussian statistics and non-Lorentzian line shape

P. Zoller, G. Alber, and R. Salvador
Theoretical Physics Institute, University of Innsbruck, A-6020 Innsbruck, Austria
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Lorentzian models for laser line shapes lead to qualitatively incorrect results for off-resonance excitation of atoms. This paper is the first attempt to present a theory of the nonperturbative interaction of an atom with a chaotic field (representing multimode laser radiation having strong amplitude fluctuations) with a line shape falling off faster than a Lorentzian. To this end we suggest a stochastic Markovian model for a non-Lorentzian chaotic field. To solve the multiplicative stochastic differential equations describing the atom-field interaction we propose a "marginal characteristic function approach." This not only reproduces our earlier results in a more elegant way and establishes the relationship between approaches used by other authors in a different context, but also provides the simplest possible basis for our present discussion of ac Stark splitting in double optical resonance. While for a chaotic field with a Lorentzian line shape the asymmetry of the two-peaked off-resonance spectrum is reversed for all values of the detuning compared with the monochromatic case, our present model predicts a reversed peak asymmetry only for detunings smaller than a few laser bandwidths in agreement with experiment. The on-resonance spectrum is dominated by the amplitude fluctuations and is only weakly affected by changes of the spectral line shape of the laser.

I. INTRODUCTION

In recent years there has been growing interest in laser temporal coherence effects in saturation and ac Stark splitting of an atomic transition.[1-13] Nearly all the work published so far on finite bandwidth excitation of atoms in intense electromagnetic fields is based on the Lorentzian line shape to model the laser spectrum. The reason for this assumption is mainly one of computational simplicity; many calculations performed so far become much more cumbersome, if not intractable, if something more complicated than a Lorentzian spectrum is assumed. Realistic laser line shapes, on the other hand, are generally expected to resemble more closely a Gaussian distribution.[13] The central question regarding the applicability of a Lorentzian profile to model a realistic laser spectrum is whether or not both lead to qualitatively different predictions. Clearly, one would expect some qualitative differences; but a qualitatively different behavior is certainly unacceptable.

The problem with Lorentzian line shapes is that the far wings fall off very slowly.[13] If we tune a radiation source with a Lorentzian spectrum many linewidths away from the resonances of an atomic system, the incoherent contributions to the excitation of this atomic state (through absorption of resonant photons out of the wing of the laser spectrum) can dominate over the coherent part for all values of the detuning. For a realistic laser spectrum with a line shape falling off much faster than a Lorentzian, these incoherent contributions should be negligible; because for large detunings one would expect the laser field to appear monochromatic to the atom. These difficulties with Lorentzian line shapes far off resonance are best illustrated by two examples.

One of the qualitatively new features, which have been observed experimentally in finite bandwidth radiation fields, is the reversed peak asymmetry in the doublet spectrum of double optical resonance (DOR) for off-resonance excitation.[13] In DOR a strongly driven atomic transition is probed by a second weak laser inducing population in a third unperturbed state. This prediction has recently been confirmed experimentally by Hogan *et al.*[13] In this experiment, however, the reversed peak asymmetry persisted only for detunings of a few laser linewidths, reverting back to normal for larger detunings, while the theory—based on the assumption of a Lorentzian line shape—predicts a reversed peak asymmetry for arbitrary detunings.[14] Physically, this reversal of the peak asymmetry is caused by the overlap of the wing of the laser spectrum with the atomic resonance, leading to an enhancement of the two-step process in comparison with the off-resonance two-photon absorption. The disagreement between theory and experiment may, therefore, be attributed to the assumption of a Lorentzian line shape in the theoretical treatment, while in the experiment the wings of the spectrum were falling off much faster than a Lorentzian. Consequently, calculations with a realistic laser spectrum are expected to lead to a reversed peak asymmetry only within a certain range of detuning. Similar problems with Lorentzian line shapes also appear in off-resonant multiphoton ionization by finite bandwidth laser light: A Lorentzian line shape of the laser field leads to incoherent excitations of the atomic levels by the photons in the wings of the laser spectrum which, even far off resonant, may dominate the ionization probability by orders of magnitude.[15] Recent experimental results, however, have demonstrated these incoherent contributions to be negligibly small.[16]

The above examples illustrate the necessity to study stochastic models with line shapes falling off faster than a Lorentzian. In recent papers we have shown how the phase-diffusion model (PDM) (describing light from a single-mode laser with stable amplitude but flucutating phase) can be generalized to include a non-Lorentzian spectrum.[9] It is the purpose of the present paper to present a theory describing the interaction of a chaotic field (CF) having a non-Lorentzian line shape with an atomic system. A CF is a radiation field with Gaussian statistics of its (complex) amplitude. It models the radiation of a multimode laser with a large number of uncorrelated modes. Only recently, it has become possible to treat rigorously the effect of amplitude fluctuations of the CF in saturation and ac Stark splitting of an atomic transition.[7,8,10,11] Owing to the difficulties in the treatment of amplitude fluctuations all the (model) calculations performed refer to Lorentzian line shapes. The present paper is, therefore, the first attempt to deal with non-Lorentzian line shapes in the presence of (strong) amplitude fluctuations.

We have decided to apply the theory to ac Stark splitting in DOR. The reasons for this choice are twofold. The experiments on ac Stark splitting in a field with strong amplitude fluctuations have so far been performed as DOR experiments.[13] Furthermore, our present results permit comparison with our earlier calculations on DOR for a Lorentzian CF and a non-Lorentzian PDM field.[9] But there is still another aspect of the present paper worth mentioning. As we have remarked before, the treatment of non-Lorentzian line shapes in the presence of amplitude fluctuations poses rather difficult mathematical problems. To solve the stochastic density matrix equations of DOR for Markovian driving fields we suggest in this paper as a new method a "marginal characteristic function approach". This will provide not only a mathematical basis as simple as possible for our present purpose, but will also show the relationship between different approaches which have been developed in the last years in a similar context. text.[1,8-10,17]

The paper is organized as follows. In Sec. II we propose a model for a non-Lorentzian chaotic field. Section III discusses the basic equations of DOR. Section IV is devoted to a discussion of multiplicative stochastic differential equations. Section V contains results obtained for DOR in a non-Lorentzian CF.

II. MODEL FOR MULTIMODE LASER RADIATION

The electric-field amplitude of a multimode laser with M modes is given by[18]

$$\epsilon(t)=\sum_{j=1}^{M}\epsilon_j e^{-i\omega_j t-i\phi_j}\,. \tag{1}$$

$\epsilon_j=(j=1,\ldots,M)$ denote the field amplitudes of the different modes, ω_j are the mode frequencies relative to a mean frequency ω, and ϕ_j is a set of randomly distributed phases, which to a good approximation are independent of each other. The mean intensity

$$I=2\epsilon_0 c\langle|\epsilon(t)|^2\rangle=2\epsilon_0 c\sum_{j=1}^{M}|\epsilon_j|^2$$

of the multimode laser radiation is the sum of the intensities of the different modes. The angular brackets $\langle\ \rangle$ denote averaging over the random phases ϕ_j. Higher-order moments of the field amplitudes are most conveniently derived from the characteristic function[19]

$$\begin{aligned}\chi_M(\lambda,\lambda)&=\langle e^{-i\lambda\epsilon(t)-i\lambda^*\epsilon^*(t)}\rangle\\&=\prod_{j=1}^{M}J_0(2|\lambda|\epsilon_j)\end{aligned} \tag{2}$$

by differentiating $\chi_M(\lambda,\lambda^*)$ with respect to λ and λ^*. J_0 is the Bessel function of zero order. In the limit of a large number of modes ($M\to\infty$, $\epsilon_j\to 0$) the characteristic function takes on the limiting form

$$\chi_\infty(\lambda,\lambda^*)=e^{-|\lambda|^2\langle|\epsilon|^2\rangle} \tag{3}$$

and the probability distribution $P(\epsilon,\epsilon^*)$ of the field amplitude, the Fourier transform of the characteristic function, becomes a Gaussian

$$\begin{aligned}P(\epsilon,\epsilon^*)&=\int\frac{d^2\lambda}{\pi^2}e^{i\lambda\epsilon+i\lambda^*\epsilon^*}\chi_M(\lambda,\lambda^*)\\&\xrightarrow{M\to\infty}\frac{1}{\pi\langle|\epsilon|^2\rangle}e^{-|\epsilon|^2/\langle|\epsilon|^2\rangle}\end{aligned} \tag{4}$$

as one expects in view of the central limit theorem.

Multitime electric-field correlation functions can be discussed in a similar way. Consider the generating functional

$$\begin{aligned}J_M(\phi,\phi^*)&=\left\langle\exp\left(-i\int\phi(t)\epsilon(t)dt-i\int\phi^*(t')\epsilon^*(t')dt'\right)\right\rangle\\&=\prod_{j=1}^{M}J_0\left\langle 2\left|\int dt\,\phi(t)\epsilon_j e^{-i\omega_j t}\right|\right\rangle,\end{aligned} \tag{5}$$

whose functional derivatives with respect to the test functions $\phi(t)$ and $\phi^*(t)$ give the field correlation functions. For the first-order correlation function we find, for example,

$$\langle\epsilon(t)\epsilon^*(t')\rangle = i^2 \frac{\delta^2}{\delta\phi(t)\delta\phi^*(t')} J_M(\phi,\phi^*)\bigg|_{\phi=\phi^*=0}$$

$$= \sum_{j=1}^{M} |\epsilon_j|^2 e^{-i\omega_j(t-t')} . \tag{6}$$

In the limit of a large number of modes, $J_M(\phi,\phi^*)$ becomes the generating functional of a (complex) Gaussian process[20]

$$J_\infty(\phi,\phi^*) = \exp\left(-\int dt\,dt'\,\phi^*(t)\langle\epsilon^*(t)\epsilon(t')\rangle\phi(t')\right) \tag{7}$$

with $\langle\epsilon(t)\epsilon(t')\rangle = \langle\epsilon^*(t)\epsilon^*(t')\rangle = 0$. Equations (4)–(7) prove in an elegant way the well-known fact that multimode laser radiation takes on Gaussian statistics, i.e., becomes an ideal CF, if the number of independent modes becomes large.[18] From Eq. (7), a CF is characterized by the infinite sequence of correlation functions[20]

$$\langle\epsilon^*(t_1)\epsilon(t_1')\cdots\epsilon^*(t_n)\epsilon(t_n')\rangle$$

$$= \sum_P \prod_{j=1}^{M} \langle\epsilon^*(t_j)\epsilon(t'_{P(j)})\rangle , \tag{8}$$

where P denotes permutation. Below we will discuss stochastic models for a CF.

Previous treatments of the interaction of an atomic system with a CF were confined to first-order correlation functions of the form[5,7,8,10]

$$\langle\epsilon^*(t)\epsilon(t')\rangle = \langle|\epsilon|^2\rangle e^{-b|t-t'|} . \tag{9}$$

The spectrum, as given by the Fourier transform of (9), is then a Lorentzian with bandwidth b. Since the time dependence of the first-order field correlation function (9) is a simple exponential, the amplitude $\epsilon(t)$ obeys a one-dimensional complex Markov process described by the Langevin equation[8,21]

$$\begin{aligned} \dot\epsilon(t) &= -b\epsilon(t) + F_\epsilon(t), \\ \dot\epsilon^*(t) &= -b\epsilon^*(t) + F_{\epsilon^*}(t) , \end{aligned} \tag{10}$$

with Gaussian random forces

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

To construct a Markovian CF with spectrum falling off faster than a Lorentzian, we generalize the correlation function (9) to a sum of two exponentials

$$\langle\epsilon(t)\epsilon^*(t')\rangle = \langle|\epsilon|^2\rangle(\beta e^{-b|t-t'|} - b e^{-\beta|t-t'|})/(\beta-b) , \tag{11}$$

so that for $\beta \gg b$ the spectrum

$$\langle|\epsilon|^2\rangle \frac{b}{\omega^2+b^2}\,\frac{\beta(\beta+b)}{\omega^2+\beta^2} \tag{12}$$

is a Lorentzian with bandwidth b, falling off as ω^{-4} for frequencies larger than the cutoff value β. It is not difficult to see that the amplitude $\epsilon(t)$ is then the projection of a two-dimensional complex Markov process[21]

$$\dot\epsilon(t) = -b\epsilon(t) + \mathfrak{F}(t) , \tag{13a}$$

$$\dot{\mathfrak{F}}(t) = -\beta\mathfrak{F}(t) + F_{\mathfrak{F}}(t) , \tag{13b}$$

with Gaussian forces

$$\langle F_{\mathfrak{F}}(t)F_{\mathfrak{F}^*}(t')\rangle = 2\langle|\epsilon|^2\rangle b\beta(b+\beta)\delta(t-t') .$$

Comparison of the Langevin equations (13a) and (10) reveals that we have replaced the δ-correlated force F_ϵ by the Gaussian force $\mathfrak{F}(t)$ with finite coherence time $1/\beta$. In general we may note that Markovian models for a CF always lead to spectral line shapes which are rational functions of ω (the correlation functions are a sum of exponentials).[21] To describe a CF with a Gaussian spectrum, for example, one would have to give up the Markov property of $\epsilon(t)$; a rigorous treatment of the interaction of an atom with a non-Markovian CF seems, however, at the moment intractable in view of its mathematical difficulties.

It is interesting at this point to compare the first-order correlation function (11) of the CF and its spectrum (12) with the modified version of the PDM, which we have suggested recently.[9] In the PDM the laser radiation is assumed to have a constant amplitude ϵ_0 but a fluctuating phase $\phi(t)\{\epsilon(t)=\epsilon_0\exp[-i\phi(t)]\}$, with $\phi(t)$ obeying the Langevin equations[9]

$$\dot\phi(t) = \mathfrak{F}_\phi(t),\ \dot{\mathfrak{F}}_\phi(t) = -\beta\mathfrak{F}_\phi(t) + F_\phi(t) . \tag{14}$$

$F_\phi(t)$ is a Gaussian random force with

$$\langle F_\phi(t)F_\phi(t')\rangle = \beta^2 2b\delta(t-t') .$$

From the first-order correlation function

$$\langle\epsilon(t)\epsilon^*(t')\rangle = \epsilon_0^2\exp\{-b[\,|t-t'| + (e^{-\beta|t-t'|}-1)/\beta]\} \tag{15}$$

we identify, assuming $\beta \gg b$, the parameter b with the bandwidth of a Lorentzian spectrum, which has a cutoff at frequency β. In the limit $\beta \gg b$ the spectrum of the PDM thus shows a qualitatively similar behavior as the spectrum of the CF as given by Eq. (12); of course, both models differ in their higher-order statistics.

III. DOUBLE OPTICAL RESONANCE

In DOR[22] we consider an atomic system with ground state $|0\rangle$ and two excited states $|1\rangle$ and $|2\rangle$, with respective energies $\hbar\omega_0 < \hbar\omega_1 < \hbar\omega_2$. The excited states $|1\rangle$ and $|2\rangle$ have natural decay widths widths κ_1 and κ_2. The first transition $|0\rangle - |1\rangle$ is assumed to be strongly driven by the CF described in Eqs. (13). 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. In order not to perturb the ac Stark splitting of the strongly driven system $|0\rangle - |1\rangle$ we adopt the weak-probe approximation, i.e., we neglect the depopulation of the two-level system $|0\rangle, |1\rangle$ by the probe laser. In the rotating-wave approximation we find the following equations for the slowly varying density matrix elements[7-9,14]:

$$\begin{aligned}
&\left(\frac{d}{dt}+\kappa_2\right)\rho_{22}(t)=i\tfrac{1}{2}\Omega'\rho_{12}(t)+\text{c.c.}\,,\\
&\left(\frac{d}{dt}+i\Delta_2+\tfrac{1}{2}\kappa_{12}\right)\rho_{12}(t)=-i\tfrac{1}{2}\Omega'\rho_{11}(t)+i\mu_{01}\epsilon(t)\rho_{02}(t)\,,\\
&\left(\frac{d}{dt}+i\Delta_1+i\Delta_2+\tfrac{1}{2}\kappa_{02}\right)\rho_{02}(t)\\
&\qquad=-i\tfrac{1}{2}\Omega'\rho_{01}(t)+i[\mu_{01}\epsilon(t)]^*\rho_{12}(t)\,,\\
&\left(\frac{d}{dt}+\kappa_1\right)\rho_{11}(t)=i\mu_{01}\epsilon(t)\rho_{01}(t)+\text{c.c.}\,,\\
&\left(\frac{d}{dt}+i\Delta_1+\tfrac{1}{2}\kappa_{01}\right)\rho_{01}(t)=i[\mu_{01}\epsilon(t)]^*[\rho_{11}(t)-\rho_{00}(t)]\,,\\
&\rho_{11}(t)+\rho_{00}(t)=1\,,
\end{aligned}\tag{16}$$

with $\kappa_{ij}=\kappa_i+\kappa_j$. μ_{01} is the dipole matrix element of the transition $|0\rangle - |1\rangle$. Ω' is the Rabi frequency of the probe. $\Delta_1=\omega-\omega_{10}$ and $\Delta_2=\omega'-\omega_{21}$ denote the detunings in the first and second transition, respectively.

Owing to the stochastic nature of the CF $\epsilon(t)$ [compare Eq. (13)], the density matrix equations (16) are a system of stochastic differential equations (SDE)[23] which must be solved for the averaged population $\langle\rho_{22}\rangle$ of level $|2\rangle$. Angular brackets $\langle\,\rangle$ denote the averaging over the flucutations of the incident field. For a CF with Lorentzian spectrum, these equations have been solved employing Fokker-Planck-eigenfunction techniques[8] and, in the strong saturation limit, by diagrammatic summation methods.[5,7] Here we are interested in solving Eqs. (16) for the non-Lorentzian CF. To this end, we suggest in the following section a "marginal characteristic function approach" to solve the stochastic differential equations (16). Readers not interested in mathematical details can proceed to Sec. V without loss of continuity.

IV. MARGINAL CHARACTERISTIC FUNCTION APPROACH TO MULTIPLICATIVE STOCHASTIC DIFFERENTIAL EQUATIONS WITH MARKOVIAN DRIVING FIELDS

In this section we study multiplicative stochastic differential equations (SDE) of the form[23]

$$\frac{d}{dt}u(t)=A[\vec{\epsilon}(t),t]u(t)\tag{17}$$

with A a matrix and $u(t)$ a column vector. $\vec{\epsilon}(t)$ denotes a set of stochastic variables. Our goal is to solve Eq. (17) for the average $\langle u(t)\rangle$ where the angular brackets denote averaging over the statistics of $\vec{\epsilon}(t)$. An equation of the form (17) arises, as we have seen, in studying the interaction of an atomic system with an intense fluctuating driving field. The difficulty in solving equations of the form (17) is that standard mathematical procedures, such as Van Kampen's cumulant expansion,[23] are perturbation expansions in terms of the parameter $\alpha\tau_c$ ($\ll 1$) where α denotes the magnitude of the fluctuations and τ_c their coherence time. The requirement of $\alpha\tau_c$ being small is, however, not generally met for the problems under consideration; in particular for the case discussed in Sec. II. If we identify α with the mean Rabi frequency $\Omega=2\mu(\langle|\epsilon|^2\rangle)^{1/2}$ and τ_c with $1/b$ or $1/\beta$, we get $\Omega/b\gg 1$ or $\Omega/\beta\cong 1$ for high intensities, respectively.

Progress in solving Eq. (17) without the assumption of a small correlation time ($\tau_c\ll 1/\alpha$) can be made if we restrict ourselves to Markov processes $\vec{\epsilon}(t)$,[19,21] for describing the fluctuations of the driving field, such as those in the model discussed in Sec. II. This assumption permits us to reformulate the problem of solving Eq. (17) for $\langle u(t)\rangle$ in terms of an equation for the so-called marginal averages $u(\vec{\epsilon},t)$.[23] Briefly, the corresponding theorem states that—given the master equation

$$\left(\frac{\partial}{\partial t}+L(\vec{\epsilon})\right)P(\vec{\epsilon},t)=0\tag{18}$$

for the probability distribution $P(\vec{\epsilon},t)$ of $\vec{\epsilon}(t)$—the required averages $\langle u(t)\rangle$ can be found by solving the equation

$$\left(\frac{\partial}{\partial t}+L(\vec{\epsilon})\right)u(\vec{\epsilon},t)=A(\vec{\epsilon},t)u(\vec{\epsilon},t)\tag{19}$$

for the marginal averages $u(\vec{\epsilon},t)$ under the initial condition $u(\vec{\epsilon},t=0)=\langle u(t=0)\rangle P_i(\vec{\epsilon},t=0)$ the initial distribution of $\vec{\epsilon}(t)$.[23] The averages $\langle u(t)\rangle$ are then given by

$$\langle u(t)\rangle=\int d\vec{\epsilon}\,u(\vec{\epsilon},t)\,.\tag{20}$$

The conventional proof of Eq. (20) involves the fact that $u(t)$ together with $\vec{\epsilon}(t)$ describes a Markov process whose joint probability distribution obeys a master equation which can be reduced to the form (19).[23] From the point of view of quantum optics this proof is somewhat abstract. In Appendix A we present a proof of Eq. (20) which is based on the assumption for the electric-field correlation functions, the quantities of direct physical significance, to have Markov property.

In the context of atom-stochastic field interaction one is particularly interested in solving the SDE (17) for Gaussian processes $\vec{\epsilon}(t)$. The electric-field amplitude $\vec{\epsilon}(t)$ of the CF and the phase $\phi(t)$ in the PDM are examples for such Gaussian Markov processes. Below we propose, with reference to the theorem quoted in Eqs. (17)–(20), a "marginal characteristic function approach" to solve the SDE (1) for a Gaussian-Markov process. Rather than presenting the method in full generality, we give an outline of this approach with several relevant examples. In particular we will apply the method to solve the stochastic density matrix equation (16) for a non-Lorentzian CF. The advantage of the method proposed here is that it gives a simpler and physically more transparent derivation of our earlier results and establishes the relationship with different methods suggested recently by other authors.

A. Real Gaussian processes

The first and simplest example we shall consider is the SDE

$$\frac{d}{dt}u(t)=[A+B\epsilon(t)]u(t)\,, \tag{21}$$

with A and B noncommuting matrices. $\epsilon(t)$ is a real Gaussian process obeying the Langevin equation[21]

$$\begin{gathered}\frac{d}{dt}\epsilon(t)=-\gamma\epsilon(t)+F(t)\,,\\ \langle F(t)F(t')\rangle=2\langle\epsilon^2\rangle\gamma\delta(t-t')\,.\end{gathered} \tag{22}$$

We have encountered an equation of this form in our work on the interaction of an atomic system with a field obeying the phase-diffusion equation (14).[9] To find the average $\langle u(t)\rangle$ we have to solve Eq. (19). In our example, $L(\epsilon)$ is given by the Fokker-Planck operator

$$L(\epsilon)=-\gamma\frac{\partial}{\partial\epsilon}\epsilon-\gamma\langle\epsilon^2\rangle\frac{\partial}{\partial\epsilon^2}\,. \tag{23}$$

The Fourier transformation of Eq. (19) which amounts to introducing the marginal characteristic function

$$u(\lambda,t)=\langle e^{-i\lambda\epsilon(t)}u(t)\rangle\equiv\int d\epsilon\, e^{-i\lambda\epsilon(t)}u(\epsilon,t) \tag{24}$$

yields the equation

$$\left(\frac{\partial}{\partial t}+\gamma\lambda\frac{\partial}{\partial\lambda}+\gamma\langle\epsilon^2\rangle\lambda^2\right)u(\lambda,t)=\left(A+iB\frac{\partial}{\partial\lambda}\right)u(\lambda,t)\,, \tag{25}$$

with $u(\lambda,t=0)=\langle u(t=0)\rangle\langle e^{-i\lambda\epsilon(t)}\rangle$, where $\langle e^{-i\lambda\epsilon(t)}\rangle=\exp(-\frac{1}{2}\lambda^2\langle\epsilon^2\rangle)$ is the characteristic function of the Gaussian process (21). Equation (25) agrees with an equation derived recently by Eberly[17] in a similar context. Eberly's starting point is an equation for the quantity $\exp[-i\lambda\epsilon(t)]u(t)$ which, with the help of the Langevin equation (22), can be written as a multiplicative stochastic differential equation, involving only the δ-correlated Gaussian variable $F(t)$:

$$\begin{aligned}&\left(\frac{d}{dt}+\gamma\lambda\frac{\partial}{\partial\lambda}+i\lambda F(t)\right)e^{-i\lambda\epsilon(t)}u(t)\\ &\qquad=\left(A+iB\frac{\partial}{\partial\lambda}\right)e^{-i\lambda\epsilon(t)}u(t)\,.\end{aligned} \tag{26}$$

Note that this reduction is possible only at the expense of introducing partial derivatives with respect to λ. The averaging over the field fluctuations of Eq. (26) can be performed by generalizing the results of Fox and Wódkiewicz.[24] The equation derived in this way is identical with (25). The equivalence of Eberly's formalism and our approach is, of course not confined to the present example but holds quite generally.

Eberly has suggested to solve Eq. (25) with a series expansion of $u(\lambda,t)$ in λ. It is not difficult to see, however, that this only reproduces a perturbation expansion for $\langle u(t)\rangle$, although in a concise way. A more appropriate way to solve (25) is an ansatz of the form

$$\begin{aligned}u(\lambda,t)&=\langle e^{-i\lambda\epsilon(t)}\rangle g(\lambda,t)\\ &=e^{-(1/2)\lambda^2\langle\epsilon^2\rangle}g(\lambda,t)\,,\end{aligned} \tag{27}$$

since $g(\lambda,t)$ can be expected to have a smoother dependence on λ than $u(\lambda,t)$. Expanding $g(\lambda,t)$ in a power series in λ,

$$g(\lambda,t)=\langle e^{(1/2)\lambda^2\langle\epsilon^2\rangle}e^{-i\lambda\epsilon(t)}u(t)\rangle=\sum_{n=0}^{\infty}[-i(\langle\epsilon^2\rangle/2)^{1/2}\lambda]^n\frac{1}{n!}\langle H_n[\epsilon(t)/(2\langle\epsilon^2\rangle)^{1/2}]u(t)\rangle \tag{28}$$

we find from (25) the infinite system of differential equations

$$\left(\frac{d}{dt}+n\lambda-A\right)\langle H_nu\rangle=(n+1)(\langle\epsilon^2\rangle/2)^{1/2}B\langle H_{n+1}u\rangle+(2\langle\epsilon^2\rangle)^{1/2}B\langle H_{n-1}u\rangle(1-\delta_{n,0}),\quad n=0,1,2,\ldots \tag{29}$$

for the one-time averages

$$\langle H_n[\epsilon(t)/(2\langle\epsilon^2\rangle)^{1/2}]u(t)\rangle\,. \tag{30}$$

Equation (29) must be solved for $\langle u(t)\rangle=\langle H_0u(t)\rangle$ under the initial condition $\langle H_nu(t=0)\rangle=\delta_{n,0}\langle u(t=0)\rangle$. Equation (29) is identical with our result in Ref. 9 which was derived from (25) by expanding the marginal averages in the complete biorthogonal set of eigenfunctions of the Fokker-Planck operator $L(\epsilon)$. The present derivation of this result has the advantages not only to be simpler but also to establish the connection between the averages (30) and the generating function (28) as well as its equation of motion (25). An explicit solution for the average $\langle u(t)\rangle$ can be constructed in terms of the Laplace inversion[9]

$$\langle u(t)\rangle=\frac{1}{2\pi i}\int ds\, e^{st}\frac{1}{s-A-\hat{K}(s)}\langle u(t=0)\rangle . \tag{31}$$

$\hat{K}(s)$ is the matrix continued fraction

$$\hat{K}(s)=B\frac{\langle\epsilon^2\rangle}{s+\gamma-A-B\dfrac{2\langle\epsilon^2\rangle}{s+\gamma-A\;\ddots}B}B . \tag{32}$$

The matrix $\kappa(\tau)=(1/2\pi i)\int ds\, e^{s\tau}\hat{K}(s)$ may be identified with the kernel in the integrodifferential equation for the average $\langle u(t)\rangle$

$$\frac{d}{dt}\langle u(t)\rangle=A\langle u(t)\rangle+\int_0^t d\tau\,\kappa(\tau)\langle u(t-\tau)\rangle . \tag{33}$$

$\kappa(\tau)$ describes the memory effects associated with the finite correlation time $1/\gamma$ of $\epsilon(t)$. In the limit of large γ, $\hat{K}(s)$ can be truncated after the first step, so that (33) takes on the form

$$\frac{d}{dt}\langle u(t)\rangle=A\langle u(t)\rangle+\langle\epsilon^2\rangle\int_0^t d\tau\, B\, e^{(-\gamma+A)\tau}B\langle u(t-\tau)\rangle . \tag{34}$$

Equation (34) is the Bourret approximation[23] for the SDE (21) and agrees with the lowest-order result (Born approximation) of the projection operator method.[25] Within the limit of validity of (34), i.e., the lowest-order term of the expansion in $\alpha\tau_c$, $\langle u(t-\tau)\rangle$ can be replaced by $\langle u(t)\rangle$ and the integral can be extended to infinity so that

$$\frac{d}{dt}\langle u(t)\rangle=\left(A+\langle\epsilon^2\rangle\int_0^\infty d\tau\, B\, e^{(-\gamma+A)\tau}B\right)\langle u(t)\rangle . \tag{35}$$

Alternatively, the first-order term of Van Kampen's cumulant expansion may be derived from (34) by the replacement $\langle u(t-\tau)\rangle\to e^{-A\tau}\langle u(t)\rangle$ to get

$$\frac{d}{dt}\langle u(t)\rangle=\left(A+\langle\epsilon^2\rangle\int_0^t d\tau\, B\, e^{(-\gamma+A)\tau}B\, e^{-A\tau}\right)\langle u(t)\rangle . \tag{36}$$

As has been pointed out by Van Kampen[23] and Terwiel,[25] (33) and (34) are equivalent to lowest order in $\alpha\tau_c$. If the matrices A and B commute, Eq. (36) is exact for arbitrary correlation time. For matrices, which commute approximately, the smallness of the noncommutivity may be used, according to the suggestion of Chaturvedi and Gardiner,[26] to generate an expansion valid for arbitrary correlation time.

B. Complex Gaussian processes

Having discussed the marginal characteristic function approach in detail for real Gaussian processes, we confine our discussion here to a brief outline of its application to complex Gaussian-Markov processes. The interaction of an atomic system with an electric field having the complex amplitude $\epsilon(t)$ leads to a stochastic density matrix equation of the form

$$\frac{d}{dt}u(t)=[A+B\epsilon(t)+C\epsilon^*(t)]u(t) , \tag{37}$$

with A, B, and C constant matrices. Again, the following steps to solve Eq. (37) for $\langle u(t)\rangle$ are easily generalized to include higher-order polynomial couplings in $\epsilon(t)$ and $\epsilon^*(t)$: Let us start our discussion by assuming $\epsilon(t)$ to be a one-dimensional complex Markov process obeying the Langevin equation (10). With the help of the Fokker-Planck operator

$$L=-b\frac{\partial}{\partial\epsilon}\;-b\frac{\partial}{\partial\epsilon^*}\epsilon^*-2b\langle|\epsilon|^2\rangle\frac{\partial}{\partial\epsilon\partial\epsilon^*} , \tag{38}$$

the Fourier transform of (19) leads to the equation

$$\left(\frac{\partial}{\partial t}+b\lambda\frac{\partial}{\partial\lambda}+b\lambda^*\frac{\partial}{\partial\lambda^*}+2b\langle|\epsilon|^2\rangle\lambda\lambda^*\right)u(\lambda,\lambda^*,t)=\left(A+Bi\frac{\partial}{\partial\lambda}+Ci\frac{\partial}{\partial\lambda^*}\right)u(\lambda,\lambda^*,t) \tag{39}$$

for the marginal characteristic function

$$u(\lambda,\lambda^*,t)=\langle e^{-i\lambda\epsilon(t)-i\lambda^*\epsilon^*(t)}u(t)\rangle . \tag{40}$$

As an ansatz, we attempt a solution of the form

$$u(\lambda,\lambda^*,t)=\langle e^{-i\lambda\epsilon(t)-i\lambda^*\epsilon^*(t)}\rangle g(\lambda,\lambda^*,t) . \tag{41}$$

Similar to Eq. (28), $g(\lambda,\lambda^*,t)$ can be expanded in a power series, regular at $\lambda=0$, in

$$\lambda=re^{-i\phi}/(\langle|\epsilon|^2\rangle)^{1/2}\text{ and }\lambda^*=re^{i\phi}/(\langle|\epsilon|^2\rangle)^{1/2},$$

$$\begin{aligned}g(\lambda,\lambda^*,t)&=\langle\exp[\lambda\lambda^*\langle|\epsilon|^2\rangle-i\lambda\epsilon(t)-i\lambda^*\epsilon^*(t)]u(t)\rangle\\&=\sum_{n=0}^\infty\sum_{\alpha=-\infty}^\infty(-i)^{|\alpha|}e^{-i\alpha\phi}\frac{r^{2n+|\alpha|}}{(n+|\alpha|)!}\\&\qquad\times\langle e^{-i\alpha\phi(t)}L_n^{|\alpha|}\left(\frac{|\epsilon(t)|^2}{\langle|\epsilon|^2\rangle}\right)u(t)\rangle ,\end{aligned}$$

with $\epsilon(t)=|\epsilon(t)|e^{-i\phi(t)}$ and L_n^α Laguerre polynomials. Inserting Eq. (42) into (39) we find an infinite system of equations for the averages

$$\langle e^{-i\alpha\phi(t)}L_n^{|\alpha|}[|\epsilon(t)|^2/\langle|\epsilon|^2\rangle]u(t)\rangle ,$$

which is identical with the one derived in our previous work[8,27] and Ref. 10. Here again we find the present method to reproduce these earlier results in a simple and elegant way.

The subject of the present requires the solution of the stochastic density matrix equation (16) with $\epsilon(t)$ the two-dimensional complex Markov process (13). The foregoing discussion to solve the SDE (37) can be immediately generalized to the present case. The only new feature is that, as a first step, we transform $\epsilon(t)$ and $\mathcal{f}(t)$ in Eq. (13) to the new set of variables

$$e=\epsilon+\mathfrak{F}/(\beta-b), \quad f=\mathfrak{F} \; . \tag{43}$$

This diagonalizes the drift terms in the Langevin equation (13). The Fourier transform of Eq. (19) then yields an equation for

$$u(\lambda,\lambda^*,\mu,\mu^*,t)=\langle\exp[-i\lambda e(t)-i\lambda^* e^*(t)-i\mu f(t)-i\mu^* f^*(t)]u(t)\rangle \; . \tag{44}$$

Defining, in analogy to Eq. (41), the quantity $g(\lambda,\lambda^*,\mu,\mu^*,t)$ by

$$u(\lambda,\lambda^*,\mu,\mu^*,t)=\langle\exp[-i\lambda e(t)-i\lambda^* e^*(t)-i\mu f(t)-i\mu^* f^*(t)]\rangle g(\lambda,\lambda^*,\mu,\mu^* t) \tag{45}$$

we derive the equation

$$\left(\frac{\partial}{\partial t}+\hat{L}\right)g=(A+iB\hat{\epsilon}+iB\hat{\epsilon}^*)g \; , \tag{46}$$

with

$$\hat{L}=b\lambda\frac{\partial}{\partial\lambda}+\beta\lambda\frac{\partial}{\partial\mu}+\text{c.c}, \tag{47a}$$

and

$$\hat{\epsilon}=\frac{\partial}{\partial\lambda}-\frac{\beta}{\beta-b}\langle|\epsilon|^2\rangle\lambda^* -\frac{1}{\beta-b}\frac{\partial}{\partial\mu}-b\langle|\epsilon|^2\rangle\mu^* \; . \tag{47b}$$

The averages $\langle u(t)\rangle$ can be found from its solution according to $g(\lambda=\cdots\mu^*=0,t)=\langle u(t)\rangle$. The details of the solution of Eq. (46), as applied to the stochastic density matrix (16), can be found in Appendix B.

V. DISCUSSION

In this section we discuss the solution of the stochastic density matrix equation (16) for DOR in the non-Lorentzian CF. Applying the methods of Sec. IV B to the system of equations (16), we find the stationary limit, when all transients have died out, that the averaged population of level $|2\rangle$, $\langle\rho_{22}(t)\rangle$, is given by

$$\langle\rho_{22}(t)\rangle=\frac{\Omega'^2}{4\kappa_1\kappa_2}\operatorname{Re}\left(4i\frac{\kappa_1}{\Omega'}\langle\rho_{12}(t)\rangle\right) . \tag{48}$$

The averaged off-diagonal density matrix element $\langle\rho_{12}\rangle$ can be found from the matrix-continued fraction

$$\begin{pmatrix}\langle\rho_{12}(t)\rangle 4i\kappa_1/\Omega' \\ \langle\rho_{11}(t)-\rho_{00}(t)\rangle\end{pmatrix}=\cfrac{1}{A_0-B_1\cfrac{1}{A_1-B_2\cfrac{1}{A_2}B_2\cdots}B_1}\begin{pmatrix}-\kappa_1\\ \kappa_1\end{pmatrix} . \tag{49}$$

A_n and B_n are two-by-two matrices. The derivation of the matrix-continued fraction solution[28] is rather lengthy; details of the mathematical steps and explicit expressions for the matrix elements of A and B are given in Appendix B. Here, we confine ourselves to a discussion of the physical contents of the solution (49). For Eq. (49) to be valid, it is necessary, that the Rabi frequency Ω is less than the curoff β ($\Omega\lesssim\beta$); but there are not any restrictions on the Rabi frequency Ω in relation to the bandwidth b. In the limit of Lorentzian laser line shape ($\beta\to\infty$), the continued-fraction solution is equivalent to our earlier results.[8]

Before discussing the influence of the laser line shape on the on-resonance ($\Delta_1=0$) spectrum of DOR (Fig. 1), let us briefly recall the basic features of on-resonance ac Stark splitting in a field with stable amplitude (PDM) and a field having amplitude fluctuations as the CF.[7,8] In an intense coherent driving field the spectrum of DOR consists of two lines which are separated by the Rabi frequency. The widths and heights of these lines are determined by the spontaneous decay constants and the bandwidth of the laser, but are independent of the light intensity. Since the splitting frequency of the doublet is proportional to the electric-field amplitude, fluctuations

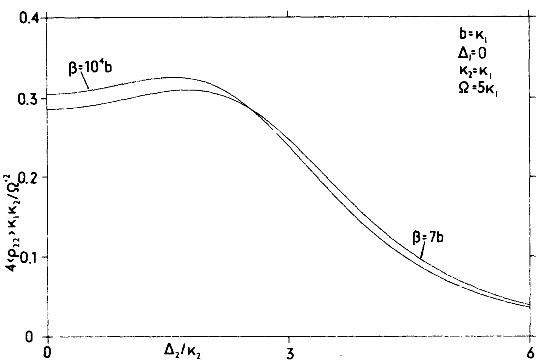

FIG. 1. The on-resonance spectrum ($\Delta_1=0$) for $\Omega=5\kappa_1$, $\kappa_1=\kappa_2=b$, and $\beta=7b,\infty$. The spectrum is symmetric around $\Delta_2=0$.

of the laser amplitude will tend to wash out the spectrum. Roughly speaking, one would expect both lines to copy the amplitude distribution of the incident light. For a CF the probability distribution of the amplitude $|\epsilon(t)|$ is

$$P(|\epsilon|)=2|\epsilon|/\langle|\epsilon|^2\rangle e^{-|\epsilon|^2/\langle|\epsilon|^2\rangle} ,$$

so that each of the lines will have a Rayleighian line shape, i.e.,

$$\langle\rho_{22}\rangle=\frac{\Omega'^2}{\kappa_2}\pi\frac{|\Delta_2|}{\Omega^2}e^{-4\Delta_2^2/\Omega^2},\quad (|\Delta_2|\gtrsim\kappa_{1,2},b) .$$

Note that in a CF the splitting frequency is reduced to $\Omega/\sqrt{2}$. With increasing light intensity the lines in the doublet are successively broadened and their height decreases.

Figure 1 shows the spectrum of DOR for on-resonance excitation ($\Delta_1=0$). The Rabi frequency was chosen as $\Omega=5\kappa_1=5b$. The two different curves correspond to $\beta=7b$ and $\beta\to\infty$ with β the cutoff of the of the laser spectrum. Note that $\beta\to\infty$ corresponds to the limit in which the lineshape of the CF becomes a Lorentzian. The line-shape dependence shows several characteristic features. A decrease of β slightly increases the splitting frequency and broadens the lines in the doublet. At the same time the heights of the peaks are reduced. This behavior is readily understood if we note that a decrease of the cutoff β increases the spectral power density of the laser spectrum at the line center. The strongly driven atom, therefore, "sees" more resonant photons. Thus, one expects the splitting frequency to be increased in comparison with the Lorentzian limit. In general we note that the high-intensity on-resonance spectrum ($\Omega\gg\kappa_{1,2},b$) is not very sensitive to the bandwidth and the line shape of the laser spectrum; the strong amplitude fluctuations of the CF are the dominating source of the broadening of the lines.

For off-resonance excitation the spectrum consists of two peaks.[29,30] In the monochromatic field the first peak corresponds (approximately) to the two-photon absorption process $|0\rangle\,\underset{\sim}{\omega}\,\underset{\sim}{\omega'}\,|2\rangle$ where the atom absorbs both a photon from the first (strong) laser and the weak-probe field. The height of this line is proportional to the intensity of the strong laser $I=2\epsilon_0\langle|\epsilon(t)|^2\rangle$ and has its maximum at $\omega'=\omega_{20}-\omega$ ($\Delta_2=-\Delta_1$). The second peak corresponds to the two-step process $|0\rangle\to|1\rangle\,\underset{\sim}{\omega'}\,|2\rangle$ where the atom makes a transition to level $|1\rangle$ by absorbing two photons from the first laser and emitting a spontaneous photon. By energy conservation this peak appears at $\omega'=\omega_{21}$ ($\Delta_2=0$). Note that this line is proportional to I^2. Therefore, in the monochromatic field the two-photon line is stronger off resonance than the two-step contribution. We call this the normal peak asymmetry. A finite bandwidth of the laser can change these features quite dramatically.[7,8] Owning to the overlap of the wing of the laser spectrum with the excited state $|1\rangle$, the two-step process $|0\rangle\to|1\rangle\,\underset{\sim}{\omega'}\,|2\rangle$ is strongly enhanced (due to the absorption of resonant photons out of the wing of the laser spectrum), while the two-photon line is merely broadened by the finite bandwidth. Thus the peak asymmetry of the DOR spectrum can be reversed in finite bandwidth excitation.[13]

To understand the line-shape dependence far off resonance, let us first concentrate on the population inversion $\langle w(t)\rangle$. Truncating the continued fraction (49) at the first step, we find

$$\langle w(t)\rangle=-\kappa_1/(\kappa_1+2W) , \tag{50}$$

with

$$W=\frac{1}{2}\Omega^2\frac{\beta}{\beta-b}\frac{\kappa_1/2+b}{\Delta_1^2+(\kappa_1/2+b)^2}-\frac{b}{\beta-b}\frac{\kappa_1/2+\beta}{\Delta_1^2+(\kappa_1/2+\beta)^2} \tag{51}$$

the induced bound-bound rate $|0\rangle-|1\rangle$. Note that W is the convolution of the spectrum of the exciting light (12) with the atomic Lorentzian, in agreement with Fermi's golden rule. While far off resonance a Lorentzian laser line shape ($\beta\to\infty$) leads to

$$W\to\frac{1}{2}\Omega^2\frac{\kappa_1/2+b}{\Delta_1^2} \tag{52}$$

neglecting terms which fall off with the fourth power of the detuning Δ_1, we find for the non-Lorentzian laser line ($\Delta_1\gg\beta$)

$$W\to\frac{1}{2}\Omega^2\frac{\kappa_1/2}{\Delta_1^2} . \tag{53}$$

The terms proportional to the bandwidth b in Eq. (52) correspond to the incoherent excitation of electrons by the wing of the laser line, while the leading term in Eq. (53) is identical with the result for a monochromatic field.

The qualitative aspects of the line-shape dependence of DOR can be understood in a similar way. Truncating again the continued fraction at the first step, the spectrum can be written in the form

$$\langle\rho_{22}\rangle = -\frac{\Omega'^2\langle w\rangle}{8\kappa_1\kappa_2}\,\frac{\Omega^2\frac{\beta}{\beta-b}\frac{1}{R_0^1}\left(\frac{\kappa_1/2+b}{R_0^{1*}}+\frac{\kappa_1/2}{T_0^1}\right)-\frac{b}{\beta-b}\frac{1}{R_0^1}\;\frac{\kappa_1/2+\beta}{R_0^{1*}}+\frac{\kappa_1/2}{T_0^1}}{i\Delta_2+\frac{1}{2}\kappa_{12}+\frac{1}{4}\Omega^2\left(\frac{\beta}{\beta-b}\frac{1}{T_0^1}-\frac{b}{\beta-b}\frac{1}{T_1^0}\right)}+\text{c.c}, \tag{54}$$

with

$$R_m^n = i\Delta_1+\tfrac{1}{2}\kappa_{10}+nb+m\beta$$

and

$$T_m^n = i(\Delta_1+\Delta_2)+\tfrac{1}{2}\kappa_{02}+nb+m\beta\ .$$

The positions and the widths of the lines of the spectrum as a function of Δ_2 are determined by the three roots of the polynomial in the denominator of Eq. (54). In the limit of well separated lines ($|\Delta_1|\gg\kappa_{1,2}$, Ω, b) we find

$$\langle\rho_{22}\rangle=\frac{1}{4}\frac{\Omega'^2}{\kappa_2}\langle\rho_{11}\rangle\left(\frac{\kappa_1}{\kappa_1/2+b}\,\frac{\beta}{\beta-b}\,\frac{\kappa_{02}/2+b}{(\Delta_1+\Delta_2)^2+(\kappa_{02}/2+b)^2}-\frac{b}{\beta-b}\,\frac{\Delta_1^2}{\Delta_1^2+\beta^2}\,\frac{\kappa_{02}/2+\beta}{(\Delta_1+\Delta_2)^2+(\kappa_{02}/2+\beta)^2}\right) \tag{55}$$

for the two-photon line ($\Delta_2\simeq-\Delta_1$) and

$$\langle\rho_{22}\rangle=\frac{1}{8}\frac{\Omega'^2}{\kappa_2}\langle\rho_{11}\rangle\left(\frac{1}{4}\frac{\Omega^2}{\Delta_1^2}+\frac{b'}{\kappa_1/2+b'}\right)\frac{\kappa_{12}/2+W/2}{\Delta_2^2+(\kappa_{12}/2+W/2)^2}\ . \tag{56}$$

for the two-step process ($\Delta_2\simeq 0$). $b'=b\beta^2/(\Delta_1^2+\beta^2)$ is a detuning-dependent effective bandwidth. The two-photon line is the convolution of the result for monochromatic excitation with the spectrum of the exciting light (12). The height of the two-step line consists of two contributions; the first, being proportional to Ω^4, is identical with the result in a monochromatic field; the second describes the incoherent population of $|1\rangle$ by the wing of the laser spectrum. For $b>\kappa_1/2$ and detunings smaller than the laser cutoff ($\Delta_1\gg\beta$), the two-step line is stronger than the two-photon process, i.e., the peak asymmetry is reversed. For $\Delta_1\gg\beta$, on the other hand, we note that the two-step line falls off with the fourth power of the detuning (Δ_1^{-4}) compared to the Δ_1^{-2} dependence of the two-photon line. With increasing detuning the two-photon line becomes stronger than the two-step process and the asymmetry reverts back to normal in agreement with the experiment of Hogan and Smith.[13] In a Lorentzian laser field the asymmetry is reversed for arbitrary detunings.

Equation (50) deserves one further comment. We derived (50) from the first-order truncation of the continued fraction (49). This is equivalent to solving the stochastic density matrix equation (16) with the help of the decorrelation ansatz[29] $\langle\epsilon(t)\epsilon(t')\rho_{ii}(t')\rangle\simeq\langle\epsilon(t)\epsilon(t')\rangle\langle\rho_{ii}(t')\rangle$. Since in the decorrelation approximation, the atom is only provided with information about the first-order electric-field correlation function, the atom is unable to distinguish between fields of different higher-order statistics. In other words, the atom "sees" the finite bandwidth of the field, but not the intensity fluctuations.[29] Consequently, the terms proportional to Ω^4 are not correctly reproduced in the approximation (50), and Eqs. (51) and (52) are almost identical with the results we have derived recently for the PDM (14).[9]

Numerical calculations on the basis of Eq. (49) confirm the above qualitative picture. Figures 2–4 compare the spectrum for different values of the detuning Δ_1 and cutoff β, similar to the on-resonance case. For larger values of the detuning (Figs. 3 and 4), a decrease of the cutoff β drastically reduces the height of the two-step line. For $\Delta_1\gg\beta$ ($\beta=5b$ in Fig. 4) the asymmetry reverses back to normal. The decrease of the intensity of the two-step process is accompanied by a slight increase of the height and a shift of the maximum of the two-photon line. This can be understood as an interference effect with the wing of the two-step line and is also due to the increase of the spectral power of the laser near the line center.

Figure 5 shows a plot of the laser spectrum together with the detuning dependence of the asymmetry parameter A.[9,13] A is defined as the difference of the heights of the two lines divided by their sum. Again we see that for detunings

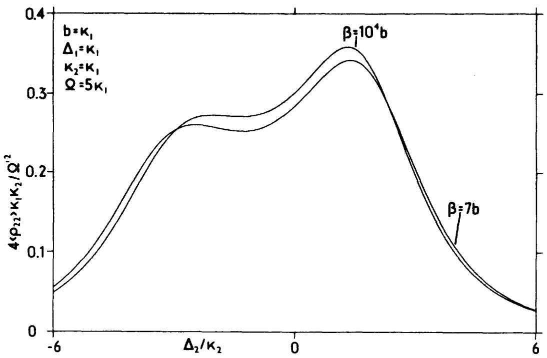

FIG. 2. The off-resonance spectrum ($\Delta_1=\kappa_1$) for $\Omega=5\kappa_1$, $\kappa_1=\kappa_2=b$, and $\beta=7b,\infty$.

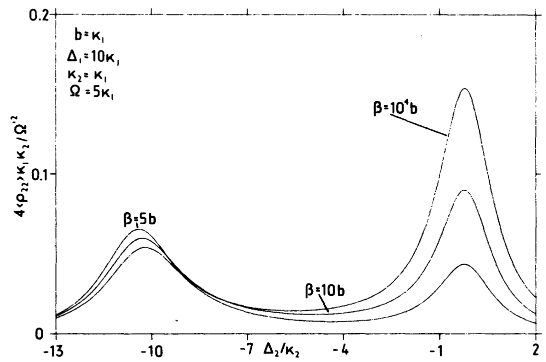

FIG. 4. The off-resonance spectrum ($\Delta_1=10\kappa_1$) for $\Omega=5\kappa_1$, $\kappa_1=\kappa_2=b$ and $\beta=5b$, $10b$, and ∞.

smaller than the cutoff, where the spectrum is essentially is Lorentzian, the asymmetry is reversed ($A<0$). As soom as the detuning Δ_1 becomes larger than β (i.e., the overlap of the wing of the laser with the resonance becomes negligible), the asymmetry reverses to normal as in the monochromatic case. The above conclustions are in qualitative agreement with the experiment of Hogan and Smith.[13] At present, a quantitative comparison with our model seems to be premature in view of the still unresolved discrepancies which have been pointed out in Ref. 7.

VI. CONCLUSIONS

In this paper we have presented a theory of the interaction of an atom with a chaotic field (representing a multimode laser field with strong Gaussian amplitude fluctuations) with a line shape falling off faster than a Lorentzian. Starting with a stochastic Markovian model for a non-Lorentzian CF. We proposed a "marginal characteristic function approach" to solve the multiplicative SDE describing the time evolution of the atom. This reproduced our earlier results for the Lorentzian CF in a more elegant way and established the relation between approaches used by other authors in a different context. It also provided a convenient basis for our discussion of ac Stark splitting in DOR in the presence of a non-Lorentzian CF.

The on-resonance doublet of DOR was shown to be dominated by the amplitude fluctuations of the CF; a decrease of the cutoff of the laser spectrum only led to a slight increase of the splitting frequency. The off-resonance spectrum turned out, however, to be extremely sensitive to the decrease of the laser spectrum in the far wings of the line. While for a CF with Lorentzian line shape the asymmetry of the spectrum of DOR is reversed for all detunings, due to the overlap of the laser line with the atomic resonance, our present model predicts a reversed asymmetry only for detunings smaller than a few laser bandwidths in agreement with experiment. The present stochastic model for a non-Lorentzian CF should also find interesting applications in related problems, as for example in multiphoton ionization.

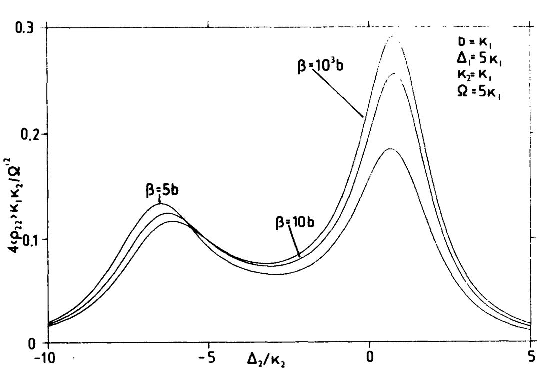

FIG. 3. The off-resonance spectrum ($\Delta_1=5\kappa_1$) for $\Omega=\kappa_1$, $\kappa_1=\kappa_2=b$ and $\beta=5b$, $10b$, and ∞.

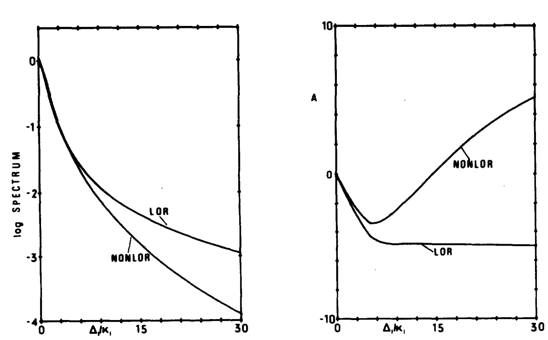

FIG. 5. The laser spectrum and the asymmetry parameter.

ACKNOWLEDGMENTS

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APPENDIX A

A formal solution of the SDE (17) for the average $\langle f(\vec{\epsilon}(t))u(t)\rangle$ with $f(\vec{\epsilon}(t))$ an arbitrary function of $\vec{\epsilon}(t)$ can be written as the perturbation series

$$\langle f(\vec{\epsilon}(t))u(t)\rangle = \langle f(\vec{\epsilon}(t))\rangle u(0) + \sum_{n=1}^{\infty}\int_0^t dt \ldots \int_0^{t_{n-1}} dt_n \left\langle f(\vec{\epsilon}(t))\prod_{j=1}^{n} A(\vec{\epsilon}(t_j),t_j)\right\rangle u(0)\,. \tag{A1}$$

For Markov processes the correlation functions on the rhs of Eq. (A1) can be expressed in terms of integrals over products of the conditional probabilities $P(\vec{\epsilon},t\,|\vec{\epsilon}\,',t')$ [with $P(\vec{\epsilon},t'\,|\vec{\epsilon}\,',t')=\delta(\vec{\epsilon}-\vec{\epsilon}\,')$] and the distribution function $P(\vec{\epsilon},t)$:

$$\begin{aligned}&\left\langle f(\vec{\epsilon}(t_0))\prod_{j=1}^{n} A((t_j),t_j)\right\rangle \\ &\quad = \int d\vec{\epsilon}_0 \ldots \int d\vec{\epsilon}_n f(\vec{\epsilon}_0)\prod_{j=1}^{n-1} P(\vec{\epsilon}_j t_j\,|\vec{\epsilon}_{j+1} t_{j+1})A(\vec{\epsilon}(t_{j+1}),t_{j+1})P(\vec{\epsilon}_n,t_n)(t_0 \geq t_1 \geq \ldots \geq t_n)\,.\end{aligned} \tag{A2}$$

Both $P(\vec{\epsilon},t\,|\vec{\epsilon}\,',t')$ and $P(\vec{\epsilon},t)$ are solutions of the master equation (18). Defining a marginal average $u(\vec{\epsilon},t)$ so that, by construction we have

$$\langle f(\vec{\epsilon}(t))u(t)\rangle = \int d\vec{\epsilon} f(\vec{\epsilon})u(\vec{\epsilon},t)\,,$$

we readily see that, in view of Eq. (A2), we can sum the perturbation series for $u(\vec{\epsilon},t)$ into the integral equation

$$u(\vec{\epsilon},t) = u(0)P(\vec{\epsilon},t) + \int_0^t dt'\int d\vec{\epsilon}\,' P(\vec{\epsilon}\,t\,|\vec{\epsilon}\,'t') \times A(\vec{\epsilon}\,',t')u(\vec{\epsilon}\,',t')\,.$$

Operating on both sides with $\partial/\partial t + L(\vec{\epsilon})$ we derive Eq. (19).

APPENDIX B

In this appendix we derive the matrix-continued-fraction solution (49) for the averages $\langle\rho_{12}\rangle$ and $\langle\rho_{11}\rangle - \langle\rho_{00}\rangle$. Defining the population inversion $W(t)=\rho_{11}(t)-\rho_{00}(t)$ in Eq. (16), the equations for the marginal characteristic functions (46) read

$$\begin{aligned}&(\partial/\partial t + L + \kappa_1)W + \kappa_1 = -2\mu\epsilon\rho_{01} + 2\mu\epsilon^*\rho_{10}\,,\\ &(\partial/\partial t + L + i\Delta_1 + \kappa_1/2)\rho_{01} = -\mu\epsilon^* W\,,\\ &(\partial/\partial t + L - i\Delta_1 + \kappa_1/2)\rho_{10} = \mu\epsilon W\,.\end{aligned} \tag{B1}$$

Expanding W, ρ_{01}, and ρ_{10} into a power series in λ and μ, $W=\sum\lambda^n\lambda^{*m}\mu^r\mu^{*s}W_{ns}^{nm}$, etc., we find in the stationary limit from Eq. (B1) an infinite system of coupled algebraic equations for W_{rs}^{nm}, $\rho_{01}{}_{rs}^{nm}$, and $\rho_{10}{}_{rs}^{nm}$. Eliminating $\rho_{01}{}_{rs}^{nm}$ and $\rho_{10}{}_{rs}^{nm}$ in these equations we derive a recursion relation for W. Confining ourselves to $\Omega<\beta$, i.e., to Rabi frequencies $\Omega=2\mu(\langle|\epsilon|^2\rangle)^{1/2}$ smaller than the cutoff of the laser spectrum, the terms W_{rs}^{nm} with $r,s\neq 0$ are seen to be small and can thus be eliminated by perturbation theory. Including terms up to $O((\Omega/\beta)^4)$ and $O(\Omega^2 b^2/\beta^4)$, we find after simple but somewhat lengthy manipulations the three-term recursion formula

$$a_{11}^n W^n = b_{11}^n W^{n-1} + b_{11}^n W^{n+1} + \kappa_1\delta_{n0}\,, \tag{B2}$$

with $W^n=[(\beta-b)/\beta]^n\langle L_n[\,|\epsilon(t)|^2/\langle|\epsilon|^2\rangle]W(t)\rangle$. The coefficients $a_{11}^n\, b_{11}^n$ are given by

$$\begin{aligned}a_{11}^n &= N_{00}^{nn} + \tfrac{1}{4}(n+1)\Omega^4 b\beta/(\beta-b)^2\, 2\,\mathrm{Re}[(D_{0\;0}^{n+1\,n} + D^{*}{}_{01}^{nn})^2]/N_{0\;1}^{n+1\,n}\\ &\quad + \tfrac{1}{4}n\Omega^4 b\beta/(\beta-b)^2\, 2\,\mathrm{Re}[(D_{0\;0}^{nn-1} + D^{*}{}_{01}^{nn})^2]/N_{0\;1}^{nn-1}\\ &\quad - \tfrac{1}{4}\Omega^4 b^2/(\beta-b)^2 (2\,\mathrm{Re}\,D_{01}^{nn})^2/N_{11}^{nn}\,,\\ b_{11}^n &= n\Omega^2\beta/(\beta-b)\{\mathrm{Re}\,D_{0\;0}^{nn-1} + \tfrac{1}{2}\Omega^2 b/(\beta-b)\,\mathrm{Re}[(D_{0\;0}^{nn-1} + D^{*}{}_{0\;1}^{nn-1})(D_{0\;0}^{nn-1} + D^{*}{}_{0\;1}^{n-1\,n-1})]\}/N_{0\;1}^{nn-1}\,,\end{aligned} \tag{B3}$$

with

$$D^{nm}_{rs} = 1/[(n+m)b + (r+s)\beta + i\Delta_1 + \kappa_1/2]$$

and

$$\begin{aligned} N^{nm}_{rs} = {} & (n+m)b + (r+s)\beta + \kappa_1 + \tfrac{1}{2}\Omega^2[\beta/(\beta-b)][(n+1)D^{n+1\,m}_{r\,s} + (m+1)D^{*\,n\,m+1}_{r\,s}] \\ & + \tfrac{1}{2}\Omega^2[\beta/(\beta-b)](m D^{n\,m-1}_{r\,s} + n D^{*\,n-1\,m}_{r\,s}) - \tfrac{1}{2}\Omega^2[b/(\beta-b)][(r+1)D^{n\,m}_{r+1\,s} + (s+1)D^{*\,nm}_{r\,s-1}] \\ & - \tfrac{1}{2}\Omega^2[b/(\beta-b)](s D^{nm}_{r\,s-1} + r D^{*\,n\,m}_{r-1\,s}) . \end{aligned}$$

The previous steps can be repeated in a similar way for the system of SDEs involving $\rho_{12}(t)$ and $\rho_{02}(t)$. After some tedious algebra, one arrives at the inhomogenous three-term recursion relation for ρ^n_{12} which, when combined with Eq. (B2), yields the matrix recursion formula

$$A_n X_n = B_n X^{n-1} + B_{n+1} X^{n+1} + Y_0 \delta_{n0} , \tag{B5}$$

with

$$X^n = \begin{pmatrix} W^n \\ 4i\kappa_1/\Omega' \rho^n_{12} \end{pmatrix}$$

and

$$Y_0 = \begin{pmatrix} -\kappa_1 \\ \kappa_1 \end{pmatrix} . \tag{B6}$$

The coefficients a^n_{22},, b^n_{22}, a^n_{21}, and b^n_{21} are given by

$$\begin{aligned} a^n_{22} = {} & \overline{N}^{nn}_{00} + \tfrac{1}{16}\Omega^4(n+1)b\beta/(\beta-b)^2/T^{n+1\,n}_{0\,0}\overline{N}^{n+1\,n}_{0\,1}T^{n+1\,n}_{0\,0} + \tfrac{1}{16}\Omega^4 nb\beta/(\beta-b)^2/T^{nn-1}_{00}\overline{N}^{nn-1}_{01}T^{nn-1}_{00} \\ & + \tfrac{1}{16}\Omega^4(n+1)b\beta/(\beta-b)^2/T^{nn}_{10}\overline{N}^{n\,n+1}_{1\,0}T^{nn}_{10} + \tfrac{1}{16}\Omega^4 nb\beta/(\beta-b)^2/T^{nn}_{10}\overline{N}^{n-1\,n}_{1\,0}T^{nn}_{10} \\ & - \tfrac{1}{16}\Omega^4 b^2/(\beta-b)^2/T^{nn}_{10}\overline{N}^{nn}_{11}T^{nn}_{10} , \end{aligned} \tag{B7}$$

$$\begin{aligned} b^n_{22} = {} & \tfrac{1}{4}n\Omega^2\beta/(\beta-b)\{1/T^{nn-1}_{00} + \tfrac{1}{4}\Omega^2[b/(\beta-b)]1/T^{nn-1}_{00}\overline{N}^{nn-1}_{01}T^{nn-1}_{00} \\ & + \tfrac{1}{4}\Omega^2[b/(\beta-b)]1/T^{nn}_{10}\overline{N}^{n-1\,n}_{1\,0}T^{n-1\,n-1}_{1\,0}\} , \end{aligned} \tag{B8}$$

$$\begin{aligned} a^n_{21} = {} & -\kappa_1 + \tfrac{1}{2}\kappa_1\Omega^2(n+1)(D^{n+1\,n}_{0\,0}/T^{n+1\,n}_{0\,0})\beta/(\beta-b)[1 + \tfrac{1}{2}\Omega b/(\beta-b)(D^{n+1\,n}_{0\,0} + D^{*\,nn}_{01})/N^{n+1\,n}_{0\,1}] \\ & + \tfrac{1}{2}\Omega^2\kappa_1 n(D^{nn-1}_{00}/T^{n\,n-1}_{0\,0})\beta/(\beta-b)[1 + \tfrac{1}{2}\Omega^2 b/(\beta-b)(D^{nn-1}_{00} + D^{*\,nn}_{01})/N^{n+1\,n}_{0\,1}] \\ & - \tfrac{1}{2}\Omega^2\kappa_1[b/(\beta-b)]D^{nn}_{10}/T^{nn}_{10} , \end{aligned} \tag{B9}$$

$$b^n_{21} = \tfrac{1}{2}\Omega^2\kappa_1 n\beta/(\beta-b)(D^{nn-1}_{00}/T^{nn-1}_{00})[1 + \tfrac{1}{2}\Omega^2 b/(\beta-b)(D^{nn-1}_{00} + D^{*\,n-1\,n-1}_{0\,1})/N^{nn-1}_{01}] , \tag{B10}$$

$$T^{nm}_{rs} = (n+m)b + (r+s)\beta + i(\Delta_1 + \Delta_2) + \tfrac{1}{2}\kappa_{02} ,$$

$$\begin{aligned} \overline{N}^{nm}_{rs} = {} & (n+m)b + (r+s)\beta + i\Delta_2 + \tfrac{1}{2}\kappa_{12} + \tfrac{1}{4}\Omega^2(n+1)[\beta/(\beta-b)]1/T^{n+1\,m}_{r\,s} + \tfrac{1}{4}\Omega^2 m[\beta/(\beta-b)]1/T^{nm-1}_{rs} \\ & - \tfrac{1}{4}\Omega^2(r+1)[b/(\beta-b)]1/T^{n\,m}_{r+1\,s} - \tfrac{1}{4}\Omega^2 s[b/(\beta-b)]/T^{nm}_{rs-1} . \end{aligned}$$

The solution of Eq. (B5) is the matrix continued fraction (49).

[1]L. D. Zusman and A. I. Burshtein, Zh. Eksp. Teor. Fiz. 61, 976 (1971) [Sov. Phys.–JETP 34, 520 (1972)].

[2]G. S. Agarwal, Phys. Rev. A 18, 1490 (1978); Z. Phys. B 33, 111 (1979).

[3]J. H. Eberly, in *Laser Spectroscopy IV*, edited by H. Walther and K. W. Rothe (Springer, Berlin, 1979), p. 80.

[4]K. Wódkiewicz, Phys. Rev. A 19, 1686 (1979).

[5]V. Yu. Finkelshtein, Zh. Eksp. Teor. Fiz. 76, 91 (1979) [Sov. Phys.–JETP 49, 46 (1979)].

[6]P. V. Elyutin, Zh. Eksp. Teor. Fiz. 69, 1156 (1975) [Sov. Phys.–JETP 42, 589 (1975)]; Opt. Spectrosc. (USSR) 43, 318 (1977); Dokl. Akad. Nauk. SSSR 235, 317 (1977) [Sov. Phys.-Dokl. 22, 390 (1977)].

[7]A. T. Georges and P. Lambropoulos, Phys. Rev. A 20, 991 (1979).

[8]P. Zoller, Phys. Rev. A 20, 1019 (1979).

[9]P. Zoller and P. Lambropoulos, J. Phys. B 12, L547 (1979); S. N. Dixit, P. Zoller, and P. Lambropoulos, Phys. Rev. A 21, 1289 (1980).

[10]A. T. Georges, Phys. Rev. A 21, 2034 (1980).

[11]M. Le Berr-Rousseau, E. Ressayre, and A. Tallet, Phys. Rev. A 22, 240 (1980).

[12]M. G. Raymer and J. Cooper, Phys. Rev. A 20, 2238

(1979).
[13]P. B. Hogan, S. J. Smith, A. T. Georges, and P. Lambropoulos, Phys. Rev. Lett. 41, 229 (1978); S. J. Smith and P. B. Hogan, in *Laser Spectroscopy IV*, edited by H. Walther and K. W. Rothe (Springer, Berlin, 1979), p. 360.
[14]A. T. Georges and P. Lambropoulos, Phys. Rev. A 18, 587 (1978).
[15]L. Armstrong and J. H. Eberly, J. Phys. B 19, L291 (1979).
[16]J. Morellec, D. Normand, G. Mainfray, and C. Manus, Phys. Rev. Lett. 44, 1394 (1980).
[17]J. H. Eberly, in *Laser Physics*, Proceedings of the Second New Zealand Summer School in Laser Physics, edited by D. F. Walls and J. D. Harvey (Academic, New York, 1980), p. 1.
[18]F. Sanchez, Nuovo Cimento 27, 305 (1975); A. V. Masalov and S. S. Todirashku, Opt. Commun. 32, 497 (1980).
[19]A. Papoulis, *Probability, Random Variables and Stochastic Processes* (McGraw-Hill, New York, 1965).
[20]R. J. Glauber, in *Quantum Optics and Electronics*, edited by C. de Witt *et al.* (Gordon and Breach, New York, 1965).
[21]M. C. Wang and G. E. Uhlenbeck, in *Noise and Stochastic Processes*, edited by N. Wax (Dover, New York, 1954).
[22]For a complete list of references see Refs. 7–9, 13, and 14.
[23]N. G. Van Kampen, Phys. Rep. 24, 171 (1976).
[24]R. Fox, J. Math. Phys. 13, 1196 (1972); K. Wódkiewicz, *ibid.* 20, 45 (1979).
[25]R. H. Terwiel, Physica (Utrecht) 74, 248 (1974).
[26]S. Chaturvedi and C. W. Gardiner (unpublished).
[27]P. Zoller, Phys. Rev. A 19, 1151 (1979); P. Zoller and P. Lambropoulos, J. Phys. B 13, 69 (1980).
[28]For a discussion of matrix-continued fractions see H. Risken and H. D. Vollmer, Z. Phys. B 39, 339 (1980).
[29]P. Zoller, in *Laser Physics*, Proceedings of the Second New Zealand Summer School in Laser Physics, edited by D. F. Walls and J. D. Harvey (Academic, New York, 1980), p. 99.
[30]In the experiment of Ref. 13 the excited-state population of level $|2\rangle$ was measured by counting the electrons which were ionized from $|2\rangle$ by the first strong laser. In this case the ionization rate is not proportional to $\langle\rho_{22}(t)\rangle$ but to $\langle|\epsilon(t)|^2\rho_{22}(t)\rangle$. This difference may be important for a quantitative interpretation of DOR experiments, but does not change the qualitative aspects of our problem regarding line-shape effects.