Non-Lorentzian laser line shapes and the reversed peak asymmetry in double optical resonance

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The interaction of an atomic system with a laser having phase fluctuations is studied within the phase-diffusion model, with the finite correlation time of the time derivative of the phase taken into account. This finite correlation time introduces a line shape which is Lorentzian near the center and falls off faster than the Lorentzian at its wings. The authors apply this model to calculate the spectrum of double optical resonance for excitation by a laser having this non-Lorentzian line shape. It is observed that the reversed peak asymmetry reverts to normal far off resonance owing to the cutoff of the laser spectrum, in agreement with recent experiments. Numerical results are presented.

I. INTRODUCTION

In recent years much work has been focused on the theoretical understanding of the excitation of an atomic transition by strong radiation of finite bandwidth.[1-10] One of the qualitatively new results has been the prediction of a reversed peak asymmetry in the doublet spectrum of double optical resonance (DOR)[6,8] and a sideband asymmetry in the triplet of resonance fluorescence (RF)[4,7] for off-resonance excitation. In DOR a strongly driven atomic transition is probed by a second, weak, laser-inducing populations to a third unperturbed state,[11,12] while in RF the fluorescence spectrum of a two-level system in an intense laser field is observed.[13] The reversed peak-asymmetry in DOR has recently been observed experimentally by Hogan *et al.*[11] In these experiments, however, the reversed peak asymmetry persisted only for detunings of a few laser linewidths, reverting to normal for larger detunings, whereas theory—based on the assumption of a Lorentzian laser line shape as given by the phase-diffusion model (PDM)—predicts a reversed peak asymmetry for arbitrary detunings.[8] Physically, the 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 to the off-resonant two-photon asborption line.[8,11] The disagreement between theory and experiment may, therefore, be attributed to the fact that the line shape of the laser (pulse) was not Lorentzian in these experiments; its wings were falling off much faster than those of a Lorentzian.[11] As a result, calculations with a realistic laser spectrum should be expected to lead to a reversed peak asymmetry only within a certain range of detunings, because for large detunings one would expect the laser to appear monochromatic to the atom.

The purpose of this paper is to present a rigorous theory of the interaction of an atom with phase-diffusing laser light of non-Lorentzian line shape. The model employed in the calculation allows for the fluctuation of not only the phase $\phi(t)$ of the field but also its time derivative $\dot{\phi}(t)$. If $\dot{\phi}(t)$ fluctuates over a time scale $1/\beta$, the wings of the resulting spectrum fall off much faster than Lorentzian for detunings larger than β. As the time $1/\beta$ tends to zero, the correlation function of $\dot{\phi}(t)$ becomes a δ function, and the spectrum tends to a Lorentzian. Thus by changing β we are able to show how certain features of the peak asymmetry in DOR arise from the long wings of the Lorentzian.

These bandwidth effects can also be given a different but equivalent interpretation by viewing the process in the time instead of the frequency domain. Knight *et al.*[7] recently articulated this interpretation, discussing the sideband asymmetry in RF in terms of what they call the "reinitiation of the transient response of the atom." This concept relies on the observation by Renaud *et al.*[14] that the off-resonant spectrum of RF becomes asymmetric if a coherent exciting field of finite duration is turned on instantaneously. This is due to the transient response of the atom, which is also known to present peculiar features in other contexts.[15] Based on this observation, Knight *et al.*[7] suggest that fluctuations of the phase $\phi(t)$ of the laser field can be viewed as continually reinforcing this transient response and thus generating asymmetric sidebands in the stationary spectrum of RF. This interpretation is consistent with the assumption of a δ-correlated time derivative $\dot{\phi}(t)$ of the phase in the PDM, which for off-resonance excitation implies fluctuations of $\dot{\phi}(t)$ over a time scale $1/\beta$ much faster than any other time scale of the problem. In particular, this means a fluctuation time $1/\beta$ much faster than $1/\Delta$, which is the time scale associated with the detuning Δ from resonance. Thus we have $1/\beta \ll 1/\Delta$ or $\beta \gg \Delta$, which of course is related to the assumption of an instantaneous pulse rise time of Renaud *et al.*[14]

In view of the recent discussion of the role of an instantaneous versus adiabatic pulse rise time in off-resonant multiphoton ionization by Theodosiou *et al.*,[15] one would expect the transient effects and therefore the sideband asymmetry in RF to be much smaller for an adiabatic turning on of the electric field far off resonance. For the same reason one expects a finite fluctuation time $1/\beta$ of $\dot{\phi}(t)$ to lead to a less pronounced sideband asymmetry for $\Delta \gg \beta$ than a δ-correlated $\phi(t)$. The same argument applies of course to the reversed peak asymmetry in DOR. The connection between the two interpretations in the time and frequency domains is established in Sec. III, where we note in detail that a finite correlation time $1/\beta$ of $\dot{\phi}(t)$ in the PDM corresponds to a cutoff of the Lorentzian laser line shape at frequencies β.

On the basis of these qualitative arguments, it is evident that far off resonance the modeling of a laser spectrum—which in its far wings falls off faster than a Lorentzian—by stochastic models with Lorentzian line shape such as the PDM is bound to lead to erroneous results. A description of the excitation of an atomic transition valid both on and far off resonance requires the study of models with non-Lorentzian line shapes. In fact, this is also supported by ideal-laser theory.[16] After presenting the theory in Sec. II, we present results in Sec. III on its application to DOR.

II. STOCHASTIC LASER MODEL

We consider a laser field with stochastic phase $\phi(t)$, constant amplitude ϵ_0, and mean frequency ω. In accordance with the laser theory of a single-mode laser far above threshold, we assume the phase to obey the Langevin equation[16]

$$d\phi(t)/dt = \mathfrak{F}(t) , \tag{1a}$$

where $\mathfrak{F}(t)$ is a Gaussian random force with correlation function

$$\langle \mathfrak{F}(t)\mathfrak{F}(t')\rangle = b\beta e^{-\beta|t-t'|} .$$

Thus $\mathfrak{F}(t)$ obeys

$$\frac{d}{dt}\mathfrak{F}(t) + \beta\mathfrak{F}(t) = F(t) , \tag{1b}$$

where F is a δ-correlated Gaussian force fulfilling

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

The phase $\phi(t)$ is therefore a projection of a two-dimensional Markov process characterized[17] by the parameters β and b. Explicit expressions for β and b in terms of fundamental laser constants are discussed by Haken.[16] The meaning of these parameters can be readily established from Eqs. (1): $1/\beta$ is the correlation time of the time derivative $\dot{\phi}(t)$ of the phase, while b gives the bandwidth of the field in the limit $\beta \to \infty$. We will consider β and b below as independent phenomenological parameters. The spectrum of the laser described by (1) is given by the Fourier transform of the correlation function:

$$\langle \exp[i\phi(t+\tau) - i\phi(t)]\rangle = \exp\{-b[|\tau| + (e^{-\beta|\tau|} - 1)/\beta]\} . \tag{2}$$

For $b \ll \beta$ the spectrum is Lorentzian with width $b_L = b$ and has a cutoff at frequencies β. In the limit $\beta \to 0$ and $b \to \infty$ with the product $(\beta b)^{1/2}$ remaining finite, the spectrum becomes Gaussian with width $b_G = (\beta b)^{1/2}$. Therefore we define, albeit somewhat arbitrarily, an effective linewidth $b_{\rm eff} = b_L b_G/(b_L + b_G)$ and a line-shape parameter $\alpha = b_L/b_G$, which for $\alpha \to 0$ gives a Lorentzian spectrum of width $b_{\rm eff} = b_L$, and which for $\alpha \to \infty$ becomes a Gaussian of width $b_{\rm eff} = b_G$.

In the limit $\beta \to \infty$, $\dot{\phi}(t)$ becomes δ correlated, and $\phi(t)$, according to (1a), obeys a Wiener-Levy[18] process, which in the context of laser theory is usually referred to as a phase-diffusion model (PDM).[16] Below we adopt the name phase-diffusion model for Eq. (1) with finite β as well.

The interaction of an atomic system with a laser whose phase is fluctuating according to the PDM leads in a natural way to the study of the multiplicative stochastic differential equation[19]

$$\left(\frac{d}{dt} + A + i\dot{\phi}(t)B\right)x(t) = 0 , \tag{3}$$

where A and B are constant matrices and $x(t)$ is a vector containing the dynamical variables of the system, whose averages $\langle x(t)\rangle$ are required. In particular, we note that, for example, the density-matrix equation and the equations for atomic correlation functions are of the type of Eq. (3).[1,10,20] Furthermore, an equation formally identical to (3) is encountered in the description of the interaction of an atom with a real Gaussian field.[21]

An equation of the form (3) has been considered by Fox,[22] van Kampen,[23] and Wódkiewicz[10] for the special case of a Gaussian δ-correlated $\dot{\phi}(t)$ ($\beta \to \infty$). They find that the averages $\langle x(t)\rangle$ obey the equation

$$\left(\frac{d}{dt} + A + bB^2\right)\langle x(t)\rangle = 0 , \quad \beta \to \infty . \tag{4}$$

We will now generalize this result for a finite correlation time of $\dot{\phi}(t) = \mathfrak{F}(t)$. To this end, we note that Eq. (3) together with (1b) describes a

Markov process.[18] The averages can therefore be found by averaging the solution of[19]

$$\left(\frac{d}{dt}+L+A+i\mathfrak{F}B\right)x(\mathfrak{F},t)=0 \tag{5}$$

according to

$$\langle x(t)\rangle=\int_{-\infty}^{+\infty}d\mathfrak{F}\,x(\mathfrak{F},t)\,. \tag{6}$$

L is the Fokker-Planck operator[17]

$$L=\beta\frac{\partial}{\partial\mathfrak{F}}+\beta^2 b\frac{\partial^2}{\partial\mathfrak{F}^2}, \tag{7}$$

of the master equation $(\partial/\partial t+L)P(\mathfrak{F},t)=0$ corresponding to the Langevin equation (1b). Equation (5) must be solved under the initial condition[19]

$$x(\mathfrak{F},t=0)=\langle x(t=0)\rangle P_0(\mathfrak{F})\,,$$

with

$$P_0(\mathfrak{F})=1/(2\beta b\pi)^{1/2}\exp(-\mathfrak{F}^2/2\beta b)$$

being the stationary distribution obeying $LP_0(\mathfrak{F})=0$. We solve Eq. (5) by expanding $x(\mathfrak{F},t)$ in the complete biorthogonal set of eigenfunctions defined by[6]

$$LP_n(\mathfrak{F})=\Lambda_n P_n(\mathfrak{F})$$

and

$$L^\dagger\phi_n(\mathfrak{F})=\Lambda_n^*\phi_n(\mathfrak{F})\,.$$

For L given by Eq. (7) the eigenfunctions with corresponding eigenvalues $\Lambda_n=n\beta$ are given by

$$P_n(\mathfrak{F})=P_0(\mathfrak{F})\phi_n(\mathfrak{F})$$

and

$$\phi_n(\mathfrak{F})=H_n\frac{[\mathfrak{F}/(2\beta b)^{1/2}]}{(2^n n!)^{1/2}}\quad(n=0,1,\ldots)\,,$$

with H_n being the Hermite polynomials. In this way the partial differential equation (5) is reduced to the infinite system of the differential equations

$$\left(\frac{d}{dt}+n\beta+A\right)x^n+iB[\beta b(n+1)]^{1/2}x^{n+1}+iB(\beta bn)^{1/2}x^{n-1}=0\quad(n=0,1,\ldots) \tag{8}$$

for the averages

$$x^n(t)=\int_{-\infty}^{+\infty}d\mathfrak{F}\,\phi_n(\mathfrak{F})x(\mathfrak{F},t)=\frac{\langle H_n[\dot\phi(t)/(2\beta b)^{1/2}]x(t)\rangle}{(2^n n!)^{1/2}}, \tag{9}$$

with

$$x^n(t=0)=\delta_{n,0}\langle x(t=0)\rangle\,.$$

Note that the averages $\langle x(t)\rangle$ are given by $\langle x(t)\rangle=x^0(t)$. Equation (8) will be the basis of our analysis of DOR in Sec. II.

The physical content of Eq. (8) valid for finite β, as opposed to that of Eq. (4), which holds in the limit $\beta\to\infty$, becomes more transparent by transforming the system (8) to an equation for the average $\langle x(t)\rangle$ alone. Taking the Laplace transform

$$\hat{x}^n(s)=\int_0^\infty dt\,e^{-st}x^n(t)$$

of Eq. (8), we find

$$\hat{x}(s)=\frac{1}{s+A+\hat{K}(s)}\langle x(t=0)\rangle\,, \tag{10}$$

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

$$\hat{K}(s)=B\frac{\beta b}{s+\beta+A+B\dfrac{2\beta b}{s+2\beta+A+\ldots}B}B\,. \tag{11}$$

Inverting the Laplace transform gives us the integro-differential equation[24]

$$\left(\frac{d}{dt}+A\right)\langle x(t)\rangle+\int_0^t K(\tau)\langle x(t-\tau)\rangle d\tau=0\,. \tag{12}$$

The kernel $K(\tau)$ clearly exhibits the memory effects associated with the finite correlation time $1/\beta$ of $\dot\phi(t)$. In the limit of rapid fluctuations of $\dot\phi(t)$ (rapid compared with the time scale associated with A), the finite correlation time of $K(\tau)$ can be ignored (Markov approximation), in which case (12) simplifies to

$$\left(\frac{d}{dt}+A+bB\frac{\beta}{\beta+A}B\right)\langle x(t)\rangle=0\,. \tag{13}$$

Equation (13) obviously reduces to Eq. (4) in the limit $\beta\to\infty$. For arbitrary correlation time, Eq. (12) or, equivalently, Eq. (8) cannot be simplified. We are able, however, to obtain an explicit exact solution for $\langle x(t)\rangle$ in the stationary limit $[dx^n(t)/dt=0]$ valid for arbitrary β. From Eq. (8) we find that the stationary average $\langle x(t)\rangle$ obeys

$$[A+K(s=0)]\langle x(t)\rangle=0\,, \tag{14}$$

where $\hat{K}(s=0)$ is the matrix-continued fraction (11). It should be noted that, in general, $\hat{K}(s=0)$ depends in a complicated way on the infinite series of atom-field correlations, as evidenced by the repeated appearance of different combinations of β, A, and B in Eq. (11).

III. APPLICATION TO DOR

We apply now to DOR the theory described in Sec. II. In DOR the Stark splitting of a strongly driven two-level atom (with the ground state denoted by $|0\rangle$, the excited state by $|1\rangle$, and their respective energies denoted by $\hbar\omega_0 < \hbar\omega_1$) is observed using a weak probe that induces a transition to another state (denoted by $|2\rangle$ with energy $\hbar\omega_2 > \hbar\omega_1 > \hbar\omega_0$).[11,26] In particular, one observes the population of level 2 as a function of the detuning of the probe field. This is usually accomplished by detecting fluorescence from $|2\rangle$ or its ionization.[11] The transitions $|0\rangle \to |1\rangle$ and $|1\rangle \to |2\rangle$ are assumed to be dipole allowed while $|0\rangle \to |2\rangle$ is assumed to be dipole forbidden. Let $E=\epsilon(e^{i\phi(t)}e^{i\omega t}+\text{c.c.})$ describe the strong exciting field, with $\phi(t)$ obeying the phase diffusion equation (1), and let $E'=\epsilon'(e^{i\omega' t}+\text{c.c.})$ describe the weak probe field, which for simplicity is assumed to be monochromatic. With the assumption that the probe field's weakness does not affect significantly the strongly driven $|0\rangle \to |1\rangle$ transition ($\rho_{22} \ll \rho_{00}, \rho_{11}$), the equations obeyed by the slowly varying density-matrix elements are[6,8,26]

$$\left(\frac{d}{dt}+k_2\right)\rho_{22}=i\tfrac{1}{2}\Omega'\rho_{12}+\text{c.c.}, \quad (15a)$$

$$\left(\frac{d}{dt}+i\Delta_2+\tfrac{1}{2}k_{12}\right)\rho_{12}=-i\tfrac{1}{2}\Omega'\rho_{11}+\tfrac{1}{2}i\Omega e^{-i\phi}\rho_{02}, \quad (15b)$$

$$\left(\frac{d}{dt}+i\Delta_1+i\Delta_2+\tfrac{1}{2}k_{02}\right)\rho_{02}=-\tfrac{1}{2}i\Omega'\rho_{01}+\tfrac{1}{2}i\Omega e^{i\phi}\rho_{12}, \quad (15c)$$

$$\left(\frac{d}{dt}+k_1\right)\rho_{11}=\tfrac{1}{2}i\Omega e^{-i\phi}\rho_{01}+\text{c.c.}, \quad (15d)$$

$$\left(\frac{d}{dt}+i\Delta_1+\tfrac{1}{2}k_{01}\right)\rho_{01}=-\tfrac{1}{2}i\Omega e^{i\phi}(\rho_{11}-\rho_{00}), \quad (15e)$$

where k_1, and k_2 are the spontaneous widths of states $|1\rangle$ and $|2\rangle$, respectively; $\Delta_1=\omega-\omega_{10}$ and $\Delta_2=\omega'-\omega_{21}$ are the detunings, and $\Omega=2\mu_{01}\epsilon$ and $\Omega'=2\mu_{12}\epsilon'$ denote the Rabi frequencies for $|0\rangle \to |1\rangle$ and $|1\rangle \to |2\rangle$ transitions, respectively, with μ_{01} and μ_{12} being the corresponding dipole moments. k_{ij} are defined by $k_{ij}=k_i+k_j$ with $k_0=0$.

The stochastic nature of the phase ϕ makes Eqs. (15) a set of stochastic differential equations.[19] One therefore needs to obtain the averaged density-matrix element $\langle\rho_{22}(t)\rangle$.

Defining new variables $\tilde{\rho}_{02}=\rho_{02}e^{-i\phi}$ and $\tilde{\rho}_{01}=\rho_{01}e^{-i\phi}$, we transform Eqs. (15) into a stochastic set of equations of the form of Eq. (3). Using Eq. (8), we find that the steady-state averages

$$\tilde{\rho}^n_{0i}=\frac{\langle H_n(\dot{\phi}(t)/(2\beta b)^{1/2})\tilde{\rho}_{0i}(t)\rangle}{(2^n n!)^{1/2}}, \quad i=1,2 \quad (16a)$$

and

$$\rho^n_{ij}=\frac{\langle H_n(\dot{\phi}(t)/(2\beta b)^{1/2})\rho_{ij}(t)\rangle}{(2^n n!)^{1/2}}, \quad i,j=1,2 \quad (16b)$$

obey

$$(k_2+\beta n)\rho^n_{22}=\tfrac{1}{2}i\Omega'\rho^n_{12}+\text{c.c.}, \quad (17a)$$

$$S_n\rho^n_{12}=\tfrac{1}{2}i\Omega\tilde{\rho}^n_{02}-\tfrac{1}{4}i\Omega' W^n-\tfrac{1}{2}i\Omega'\delta_{n,0}, \quad (17b)$$

$$T_n\tilde{\rho}^n_{02}+i[b\beta(n+1)]^{1/2}\tilde{\rho}^{n+1}_{02}+i(b\beta n)^{1/2}\tilde{\rho}^{n-1}_{02} -\tfrac{1}{2}i\Omega\rho^n_{12}=-\tfrac{1}{2}i\Omega'\tilde{\rho}^n_{01}, \quad (17c)$$

$$B_nW^n+i\Omega(\tilde{\rho}^n_{10}-\tilde{\rho}^n_{01})=-k_1\delta_{n,0}, \quad (17d)$$

$$R_n\tilde{\rho}^n_{01}+i[b\beta(n+1)]^{1/2}\tilde{\rho}^{n+1}_{01}+i(b\beta n)^{1/2}\tilde{\rho}^{n-1}_{01} -\tfrac{1}{2}i\Omega W^n=0, \quad (17e)$$

where $B_n=k_1+\beta n$, $R_n=i\Delta_1+\frac{1}{2}k_{01}+n\beta$, $S_n=i\Delta_2+\frac{1}{2}k_{12}+n\beta$, $T_n=i\Delta_1+i\Delta_2+\frac{1}{2}k_{02}+n\beta$, and $W^n=\rho^n_{11}-\rho^n_{00}$; we have also made use of the relation $\rho^n_{11}+\rho^n_{00}=\delta_{n,0}$.

Our objective is to calculate $\rho^0_{22}=\langle\rho_{22}(t)\rangle$, since the observed signal in DOR is proportional to the population of the level 2 $\langle\rho_{22}(t)\rangle$ (Ref. 11). From (17a) this is given by

$$\langle\rho_{22}(t)\rangle=\rho^0_{22}=\tfrac{1}{2}i\Omega'\rho^0_{12}/k_2+\text{c. c.} \quad (18)$$

Eliminating ρ^n_{12} and W^n from Eqs. (17b)–(17e), we obtain

$$(T_n+\tfrac{1}{4}\Omega^2/S_n)\tilde{\rho}^n_{02}+i[b\beta(n+1)]^{1/2}\tilde{\rho}^{n+1}_{02} +i(b\beta n)^{1/2}\tilde{\rho}^{n-1}_{02}+\tfrac{1}{2}i\Omega'(1-\tfrac{1}{4}\Omega^2/S_nB_n)\tilde{\rho}^n_{01} +\tfrac{1}{8}i(\Omega'\Omega^2/S_nB_n)\tilde{\rho}^n_{10}=0, \quad (19a)$$

$$(R_n+\tfrac{1}{2}\Omega^2/B_n)\tilde{\rho}^n_{01}+i[b\beta(n+1)]^{1/2}\tilde{\rho}^{n+1}_{01} +i(b\beta n)^{1/2}\tilde{\rho}^{n-1}_{01}-\tfrac{1}{2}(\Omega^2/B_n)\tilde{\rho}^n_{10}=-\tfrac{1}{2}i\Omega\delta_{n,0}, \quad (19b)$$

and taking the complex conjugate of Eq. (19b), we obtain the equation for $\tilde{\rho}^n_{10}$, viz.,

$$(R^*_n+\tfrac{1}{2}\Omega^2/B_n)\tilde{\rho}^n_{10}-i[b\beta(n+1)]^{1/2}\tilde{\rho}^{n+1}_{10} -i(b\beta n)^{1/2}\tilde{\rho}^{n-1}_{10}-\tfrac{1}{2}(\Omega^2/B_n)\tilde{\rho}^n_{01}=\tfrac{1}{2}i\Omega\delta_{n,0}. \quad (19c)$$

Defining

$$x^n=\begin{pmatrix}\tilde{\rho}^n_{10}\\ \tilde{\rho}^n_{01}\\ \tilde{\rho}^n_{02}\end{pmatrix}, \quad y_0=\tfrac{1}{2}i\Omega\begin{pmatrix}1\\1\\0\end{pmatrix}, \quad (20)$$

and $a_n=b\beta n$, we can rewrite Eq. (19) as

$$V_nx^n-i(a_{n+1})^{1/2}x^{n+1}-i(a_n)^{1/2}x^{n-1} =y_0\delta_{n,0} \quad (n=0,1,\ldots), \quad (21)$$

where the matrix V_n is given by

$$V_n=\begin{pmatrix} R_n^* + \frac{1}{2}\Omega^2/B_n & -\frac{1}{2}\Omega^2/B_n & 0 \\ \frac{1}{2}\Omega^2/B_n & -(R+\frac{1}{2}\Omega^2/B_n) & 0 \\ -\frac{1}{8}i\Omega'\Omega^2/S_nB_n & -i\Omega'(1-\frac{1}{4}\Omega^2/S_nB_n) & -(T_n+\frac{1}{4}\Omega^2/S_n) \end{pmatrix}. \tag{22}$$

The solution of (21) for x^0 can be expressed in the form of a matrix-continued fraction:

$$x^0=\begin{pmatrix}\tilde{\rho}^0_{10}\\ \tilde{\rho}^0_{01}\\ \tilde{\rho}^0_{02}\end{pmatrix}=\begin{pmatrix}\langle\tilde{\rho}_{10}(t)\rangle\\ \langle\tilde{\rho}_{01}(t)\rangle\\ \langle\tilde{\rho}_{02}(t)\rangle\end{pmatrix} = \cfrac{1}{V_0+\cfrac{a_1}{V_1+\cfrac{a_2}{V_2+\cfrac{a_3}{V_3+\dots}}}}\,y_0\,, \tag{23}$$

analogous to Eq. (14). Since the matrices V_n are nonsingular, there is no ambiguity in the form of the solution in Eq. (23). Even though there seem to be no general theorems on the convergence of matrix-continued fractions, numerical calculations indicate that convergence is achieved with only a few terms for a wide range of the parameters. For smaller values of β the convergence is the worst, while for $\beta\to\infty$ immediate convergence is obtained after the first term.

Having calculated $\langle\tilde{\rho}_{10}(t)\rangle$, $\langle\tilde{\rho}_{01}(t)\rangle$, and $\langle\tilde{\rho}_{02}(t)\rangle$ from (23), one can proceed to calculate $\langle\rho_{22}(t)\rangle$ with the help of Eqs. (17a), (17b), and (17d), with $n=0$. This is done numerically, and typical results are presented in Sec. IV.

IV. NUMERICAL CALCULATIONS AND RESULTS

We have calculated $\langle\rho_{22}(t)\rangle$, as described in Sec. III. The parameters chosen in these calculations were $k_1=k_2=1.0$, $\Omega=4.0$, $\Omega'=0.4$, $\beta=10$, and $b=1.0$. This corresponds to the line-shape parameter $\alpha=1/\sqrt{10}$ and the effective bandwidth $b_{\text{eff}}=\sqrt{10}/(1+\sqrt{10})\approx 1$. For a fixed value of Δ_1, $\langle\rho_{22}(t)\rangle$ is plotted as a function of Δ_2 in Figs. 1 and 2. When $\Omega\ll k_1$, $\langle\rho_{22}(t)\rangle$ exhibits a peak around $\Delta_1+\Delta_2\approx 0$, while for $\Omega\gg k_1$ the spectrum becomes a doublet owing to Stark splitting. One of the peaks occurs at $\Delta_1+\Delta_2\approx 0$ and corresponds to the off-resonant absorption of a photon from each laser to populate level 2 ($|0\rangle\xrightarrow{\omega\,\omega'}|2\rangle$) (Ref. 8). We call this the "two-photon" process. The other peak, which occurs near $\Delta_2\approx 0$, corresponds to the "two-step" excitation $|0\rangle\xrightarrow{\omega}|1\rangle\xrightarrow{\omega'}|2\rangle$ (Ref. 8). For a monochromatic exciting laser ($b=0$) the two-photon line is proportional to the intensity of the strong laser, while the two-step line is proportional to the square of the intensity. Therefore, in the case of monochromatic fields[8] the two-photon line will be stronger than the two-step line. This is known as the normal peak asymmetry.

Let h_U and h_L correspond to the heights of the peaks of $\langle\rho_{22}(t)\rangle$ which (for fixed Δ_1) occur for larger and smaller values of Δ_2, respectively. Then h_U and h_L correspond to the two-step and two-photon processes, respectively, for $\Delta_1>0$ and vice versa for $\Delta_1<0$. Thus with a monochromatic exciting laser $h_U<h_L$ for $\Delta_1>0$, while $h_U>h_L$ for $\Delta_1<0$. Defining the asymmetry parameter[11] A as

$$A=(h_L-h_U)/(h_L+h_U)\,, \tag{24}$$

we clearly see that $A>0$ for $\Delta_1>0$, and $A<0$ for $\Delta_1<0$ correspond to normal asymmetry.[25]

When the exciting field has a finite bandwidth, the two-step process is enhanced by what can be

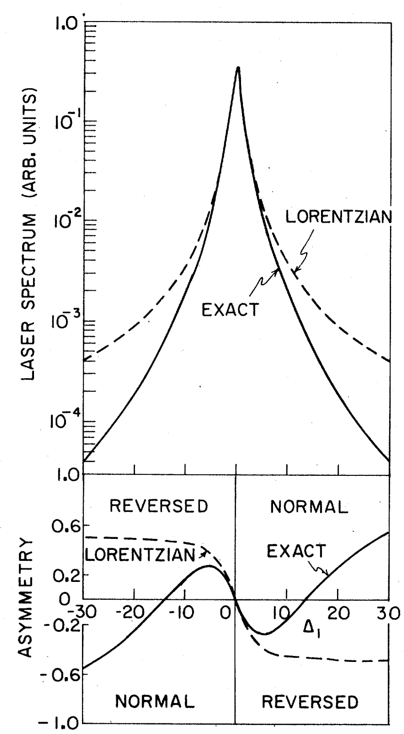

FIG. 1. Laser spectrum and asymmetry parameter A for DOR. "Exact" corresponds to the spectrum given by Eq. (25) for $b=1$ and $\beta=10$, "Lorentzian" to the case $b=1$ and $\beta\to\infty$. $\Omega=4.0$, $\Omega'=0.4$, $k_1=k_2=1$, $b=1$, and $\beta=10$ are used in the calculation of the asymmetry for the exact case, while $\beta=\infty$ with the other parameters fixed is used for the Lorentzian case.

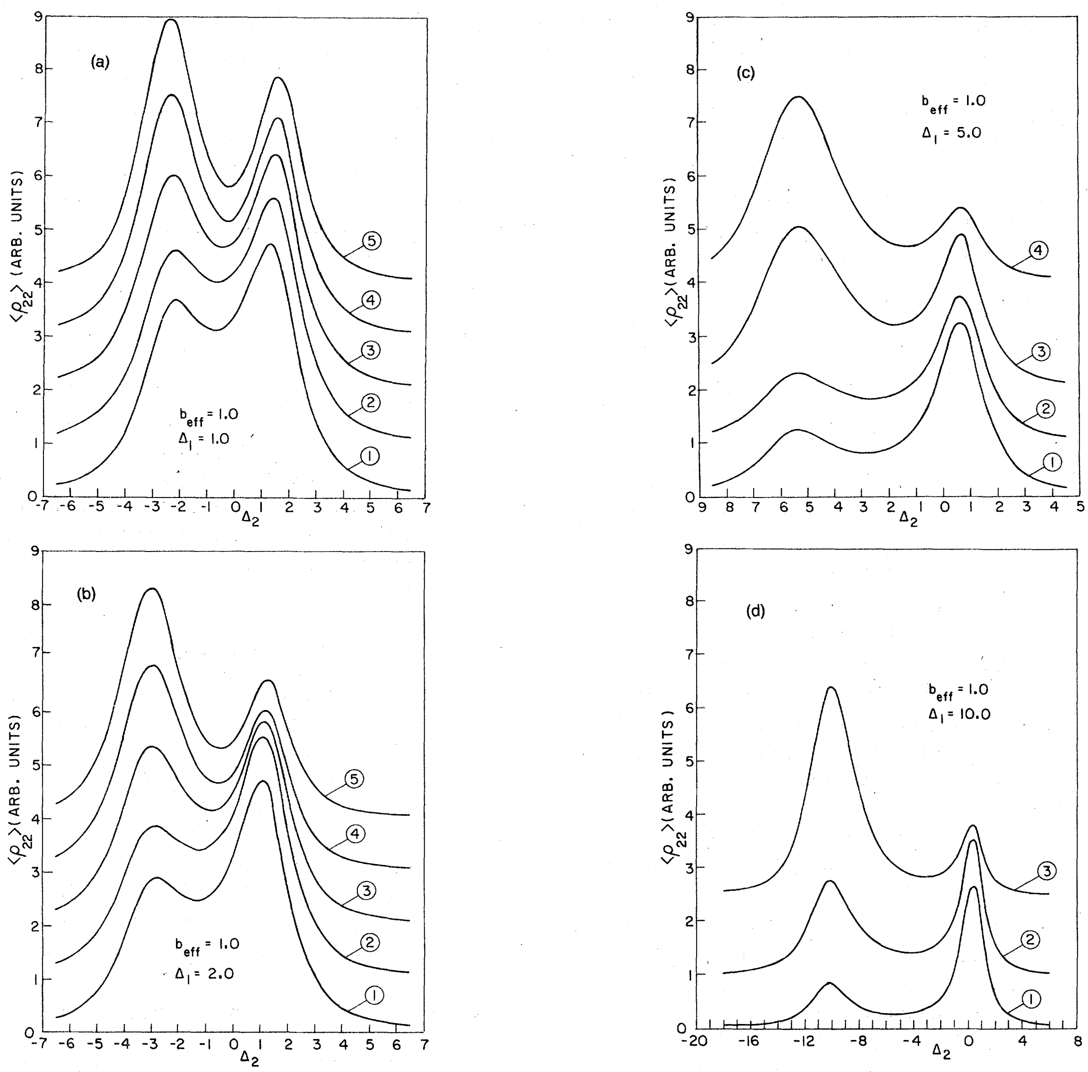

FIG. 2. DOR spectrum $\langle\rho_{22}(t)\rangle$ as a function of Δ_2 for various values of Δ_1 and α. $l=4.0$, $\Omega'=0.4$, $k_1=k_2=1$, and $b_{eff}=1$ for all curves. (a)–(d) correspond to $\Delta_1=1.0$, 2.0, 5.0, and 10.0, respectively; in each, curves 1–5 correspond to $\alpha=0.2$, 0.5, 1.0, 2.0, and 5.0, respectively. For clarity, each of the curves is shifted by a constant with respect to the previous one.

described qualitatively as the absorption of photons from the wings of the laser. This will generally cause the asymmetry to be reversed, since the two-step process becomes resonant while the two-photon process is still off resonant. Therefore, for finite-bandwidth lasers with Lorentzian line shapes, we have $A<0$ for $\Delta_1>0$ and $A>0$ for $\Delta_1<0$. This has been demonstrated explicitly in recent theoretical work.[8]

Now if the spectrum of the exciting laser falls off faster than the Lorentzian, the absorption from the wings of the spectrum will weaken thereby reducing the enhancement of the two-step process. Thus if the spectrum falls off sufficiently fast, one should expect the asymmetry to revert to normal, viz., $A>0$ for $\Delta_1>0$ and $A<0$ for $\Delta_1<0$. This behavior is clearly demonstrated by the laser model in Fig. 1.

The laser spectrum for the phase-diffusion model described in Sec. I is proportional to

$$I \sim \mathrm{Re}\left[\frac{e^{b/\beta}}{\beta}\left(\frac{\beta}{b}\right)^{(b-i\Delta)/\beta}\gamma\left(\frac{b-i\Delta}{\beta},\frac{b}{\beta}\right)\right], \qquad (25)$$

with γ being the incomplete γ function, and is plotted in the upper half of Fig. 1 for $b=1$ and $\beta=10$. The corresponding asymmetry parameters designated as "exact" for $b=1$, and $\beta=10$ and "Lorentzian" for $b=1$ and $\beta\to\infty$, are plotted in the lower half. The Lorentzian spectrum with $b=1$ is also plotted for comparison.

As is evident from Fig. 1, the asymmetry is reversed for $\Delta_1 \ll \beta$. For $\Delta_1 \gg \beta$ the asymmetry reverts to normal for the exact case, while it stays reversed for the Lorentzian case, in agreement with our preliminary and more qualitative discussion in Ref. 20. The reversal of the asymmetry to normal for larger detunings is explained by the fact that the two-step process excited by the wings of the spectrum is weaker owing to the cutoff of the laser line shape. The asymptotic values of A are ± 1 for finite values of β, while for the Lorentzian line shape ($\beta \to \infty$).

$$A \to \pm \frac{(k_1+k_2)k_1 - 2b(2b+k_2)}{k_1(k_1+k_2)+2b(2b+k_2)}, \qquad (26)$$

$$\Delta_1 \gtrless 0, \quad |\Delta_1| \gg k_1, \ 2b, \ k_2 .$$

For $k_1 = k_2 = 1$ and $b = 1$, $A \to \pm 0.5$, as seen in Fig. 1. Because the present model does not incorporate intensity fluctuations, these results for the exact case cannot be compared quantitatively with the experimental results of Hogen *et al.*,[11] since the laser they used had some intensity fluctuations as well.

In Fig. 2 we plot $\langle \rho_{22} \rangle$ for various values of the line-shape parameters α. Recall that for $\alpha \to 0$ the line shape becomes Lorentzian, whereas for $\alpha \to \infty$ the line shape assumes a Gaussian form. Again we have chosen $\Omega = 4.0$, $\Omega' = 0.4$, $k_1 = 1 = k_2$, and $b_{\text{eff}} = 1.0$ as in Fig. 1. In all parts [(a)–(d)], curves 1–5 correspond to $\alpha = 0.2$, 0.5, 1.0, 2.0, and 5.0, respectively. As can be seen from Figs. 1 and 2, for smaller values of α the asymmetry of the spectrum is reversed, while increasing values of α cause the spectrum to revert to normal for a fixed value of Δ_1. The particular value of α at which the asymmetry reverts to normal depends on the value of Δ_1, since the more physical comparison is whether $\Delta_1 \gg \beta$ or $\Delta_1 \ll \beta$. The curves are plotted for $\Delta_1 = 1.0$, 2.0, 5.0, and 10.0 and exhibit the reversal to normal asymmetry for smaller values of α an Δ_1 is increased. For clarity, each of these curves has been shifted by a constant in the vertical direction relative to the previous one.

In conclusion, we have demonstrated that within the PDM model described in Sec. I the asymmetry in the DOR spectrum reverts to normal far off resonance when the wings of the laser spectrum fall off faster than the Lorentzian. The atom sees the field as monochromatic far away from the center of the spectrum owing to the cutoff of the spectrum described by Eq. (1). Since in practice the laser spectrum is Lorentzian only for a few laser linewdiths, the calculations performed with the present model should have many applications. For example, some of the results presented in the literature for multiphoton ionization with a Lorentzian spectrum would be affected by the non-Lorentzian line shapes of the laser. Such a calculation based on the present model will be presented elsewhere.

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APPENDIX

The usual method of calculating a continued fraction (CF) has been to truncate the CF and evaluate it starting from below. However, this method is cumbersome and time consuming for checking the convergence of the matrix-continued fraction (MCF). In this appendix we generalize a standard technique for calculating ordinary CF's to MCF's.[26] This allows one to calculate the MCF starting from the top in successive approximations with a minimum number of matrix operations.

Consider a three-term recursion relation

$$c_p X_p + d_p X_{p+1} + e_p X_{p-1} = \delta_{p,0} Y_0 \quad (p = 0, 1, \ldots), \qquad \text{(A1)}$$

where c_p, d_p, and e_p ($e_0 = 0$) are $n\times n$ matrices and X_p and Y_0 are n-dimensional column vectors. Equation (A1) must be solved for X_0. Writing $X_p = Z_p d_0^{-1} Y_0$ with Z_p $n\times n$ matrices we see that, the Z_p's fulfill

$$Z_{p+1} = -b_{p+1} Z_p + a_p Z_{p-1} + \delta_{p0} 1 \quad (p = 0, 1, \ldots), \qquad \text{(A2)}$$

where $a_p = -d_p^{-1} e_p$ and $b_p = d_{p-1}^{-1} c_{p-1}$ ($p = 1, 2, \ldots$). Equation (A3) has the explicit solution for Z_0,

$$Z_0 = \cfrac{1}{b_1 + \cfrac{1}{b_2 + \ldots} a_2} a_1 . \qquad \text{(A3)}$$

Successive approximations of (A3) can be found from

$$Z_0^{(r)} = B_r^{-1} A_r \quad (r = 0, 1, \ldots), \qquad \text{(A4)}$$

in the sense that

$$Z_0 = \lim_{r\to\infty} Z_0^{(r)} . \qquad \text{(A5)}$$

The matrices A_p and B_p can be recursively calculated from

$$\left.\begin{aligned} A_{p+1} &= b_{p+1}A_p + a_{p+1}A_{p-1} \\ B_{p+1} &= b_{p+1}B_p + a_{p+1}B_{p-1} \end{aligned}\right\} \quad (p=0,1,\dots), \qquad \text{(A6)}$$

with the starting values $A_{-1}=1$, $A_0=0$, $B_{-1}=0$, and $B_0=1$. Equation (A4) together with (A6) constitutes the desired set of equations. Equations (A5) and (A6) can be easily proved by induction. We have applied this procedure in calculating the MCF given in this paper.

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$$A = (h_{\rm TP} - h_{\rm TS})/h_{\rm TP} + h_{\rm TS}),$$

where $h_{\rm TP}$ and $h_{\rm TS}$ are the heights of the two-photon and two-step peaks. With this definition we would have $A < 0$ for reversed asymmetry and $A > 0$ for normal, irrespective of the sign Δ_1. In this paper, however, we use A as defined in Eq. 24 in order to be consistent with the definition of Hogan *et al.* (Ref. 11).
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