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Saturation of two-level atoms in chaotic fields

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Exact solutions are presented for the equations describing the stationary population of a two-level atom in a nonmonochromatic ideal chaotic field. The dependence of the excited-state population on the Rabi frequency, bandwidth of the field, spontaneous decay, and detuning is studied and compared with the results obtained for phase-diffusing laser light. The validity of the decorrelation approximation is also investigated.

I. INTRODUCTION

During the last two years, increasing interest has focused on the problem of resonant atomic transitions saturated under stochastically fluctuating nonmonochromatic light sources. Its aspects have been discussed in various contexts such as resonance fluorescence,[1-6] ac Stark splitting in double optical resonance[7-10] and multiphoton ionization.[11-14] The theory of these effects has been based on the decorrelation approximation[12,13] (DA) which is rigorously valid for light with perfectly stable amplitude but a diffusing phase,[1,5,9] as is the case with a stabilized well-above-threshold cw single-mode laser. Under these conditions, the effect of the finite laser bandwidth is well understood and some of the theoretical predictions are in agreement with recent experimental results.[15] The understanding of these effects under chaotic fields (CF) is, however, very incomplete. Relevant discussions have been either purely qualitative[4] or have employed the DA[12,13] whose validity in the regime of saturation under a CF is at best doubtful, unless the laser bandwidth is much larger than other relaxation parameters (widths).[12,14] On the other hand, experiments with pulsed multimode lasers correspond much more closely to the CF case. There is in fact some evidence that some of its effects may have been seen in a recent experiment.[15] It is therefore necessary to gain quantitative understanding of saturation under chaotic fields. In addition to its relevance to the interpretation of experiments, such understanding provides valuable insight into the nature of the interaction of strong fields with atomic and molecular systems.

In recent work, we have introduced a formalism capable of dealing with a chaotic as well as a phase diffusing field.[5,14] The chaotic field with a nonzero bandwidth introduces considerable mathematical complications which render exact solutions extremely difficult if possible at all. Thus the few cases for which exact solutions can be found[7,8,10] acquire particular significance in this context as they can provide a guide for the understanding of more complicated cases. Moreover such exact solutions are important in their own right since they contain most of the basic phenomena of the more general problem.

In this paper I present an exact solution for the stationary population of a two-level atom (TLA) strongly driven by a nonmonochromatic ideal CF. These stationary populations can be measured, for example, by monitoring the total fluorescence from the excited state of the TLA. Furthermore, knowledge of these stationary density-matrix elements is required in the more complicated theoretical calculations of resonance fluorescence and ac Stark splitting in double-resonance experiments. The TLA is the basic ingredient of saturation studies and although the presence of a probe—as in double resonance—perturbs the stationary state, this perturbation does not destroy the basic features of the exact results, provided the probe is sufficiently weak. In Sec. II, we introduce our model of an ideal CF. Starting from stochastic optical Bloch equations, we derive a continued fraction expansion for the stationary density matrix elements. In the third section we study the dependence of the excited state population on the Rabi frequency, bandwidth of the CF, and detuning. Also we compare it with results obtained by the decorrelation approximation or equivalently the solution for purely phase-diffusing laser light.

II. MODEL FOR ATOM-CHAOTIC FIELD INTERACTION

For the CF we adopt the simple model[14] in which the complex amplitude of the field $\epsilon(t)$ obeys the Langevin equations

$$\dot{\epsilon}(t) = -b\epsilon(t) + F_\epsilon(t), \quad \dot{\epsilon}^*(t) = -b\epsilon^*(t) + F_{\epsilon^*}(t), \quad (1)$$

with Gaussian random forces

$$\langle F_\epsilon(t) F_{\epsilon^*}(t')\rangle = 2b\langle|\epsilon|^2\rangle\delta(t-t'),$$
$$\langle F_\epsilon(t)F_\epsilon(t')\rangle = \langle F_{\epsilon^*}(t)F_{\epsilon^*}(t')\rangle = 0.$$

Thus, $\epsilon(t)$ obeys a normal Markov process.[16] From the first-order correlation function

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

where b and $\langle|\epsilon|^2\rangle$ are identified with the bandwidth of the Lorentzian spectrum and the expectation value $\langle|\epsilon(t)|^2\rangle$ of the electric field, respectively. As a consequence of the Gaussian property of $F_\epsilon(t)$ and $F_{\epsilon^*}(t)$, a higher-order correlation function fulfills the equation

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

thus confirming that $\epsilon(t)$ is an ideal chaotic field.[17]

The time evolution of the strongly driven TLA is determined by the optical Bloch equations[18] which under the presence of a stochastic driving field become a system of stochastic differential equations.[16] The usual way to solve these equations for nonmonochromatic fields has been the DA which decorrelates atom-field averages[2,3,12,13]

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

where $\rho_{ij}(t)$ denote the density matrix elements of the TLA. This DA, although perfectly correct for the phase diffusion model,[1,5,9] can be justified in general, and specifically for the CF described above only in the limit where the Rabi frequency $\Omega = 2\mu(\langle|\epsilon|^2\rangle)^{1/2}$—with μ the dipole matrix element of the TLA—is much smaller than the bandwidth b, or the spontaneous decay rate κ or the detuning Δ of the mean frequency of the laser from the resonance frequency of the TLA. Thus, in general, the DA becomes questionable if the driving field is sufficiently intense to saturate the transition in the TLA. In this approximation, which we indicate by a subscript DA in the average, we find for the averaged stationary population inversion

$$\langle W(t)\rangle_{\rm DA} = \langle \rho_{11}(t)-\rho_{00}(t)\rangle = \frac{-\kappa}{\kappa+\Omega^2[\frac{1}{2}\kappa+b]/[\Delta^2+(\frac{1}{2}\kappa+b)^2]} . \tag{3}$$

Note that the same result is obtained for the phase diffusion model[3] which has the same spectrum as our CF but differs in higher-order correlations.[19] For the phase-diffusion model, however, (3) is exact.

As we have shown recently,[14] the stochastic optical Bloch equations can be reduced to an infinite set of differential equations for certain one-time atom-field averages:

$$i\left(\frac{d}{dt}+\kappa+2bn\right)W^n - \Omega(n+1)^{1/2}\rho_-^n + \Omega\sqrt{n}\,\rho_-^{n-1} + i\kappa\delta_{n,0} = 0 , \tag{4a}$$

$$i\left(\frac{d}{dt}+\frac{\kappa}{2}+(2n+1)b\right)\rho_-^n + \Delta\rho_+^n - \Omega(n+1)^{1/2}W^n + \Omega(n+1)^{1/2}W^{n+1} = 0 , \tag{4b}$$

$$i\left(\frac{d}{dt}+\frac{\kappa}{2}+b(2n+1)\right)\rho_+^n + \Delta\rho_-^n = 0 ,\quad n=0,1,\ldots . \tag{4c}$$

W^n and $\rho_\pm^n$ are defined by

$$W^n = \langle L_n(|\epsilon(t)|^2/\langle|\epsilon|^2\rangle)(\rho_{11}(t)-\rho_{00}(t))\rangle = \rho_{11}^n - \rho_{00}^n , \tag{5a}$$

$$\rho_\pm^n = \langle \epsilon^*(t)L_n^1(|\epsilon(t)|^2/\langle|\epsilon|^2\rangle)\rho_{10}(t)\rangle/[(n+1)^{1/2}\langle|\epsilon|\rangle] \pm \text{c.c.} \tag{5b}$$

with $L_n^\alpha(x)$ being the Laguerre polynomials.[20] Thus for $n=0$, we identify $w^0 = \langle w(t)\rangle = \langle\rho_{11}(t)-\rho_{00}(t)\rangle$ with the averaged population inversion of the TLA.

Restricting ourselves to the stationary limit of the averaged atomic density matrix, Eqs. (4) become an infinite system of linear algebraic equations. Eliminating ρ_+^n and ρ_-^n in favor of W^n, we find a three-term recursion formula for W^n,

$$W^n - a_{n+1}W^{n+1} - b_n W^{n-1} = \langle W(t)\rangle_{\rm DA}\,\delta_{n,0} , \tag{6}$$

with the coefficients given by

$$a_n = \frac{A_n}{\kappa+2(n-1)b+A_n+A_{n-1}} ,\quad b_n = \frac{A_n}{\kappa+2nb+A_{n+1}+A_n} ,\quad A_n = n\Omega^2\frac{\frac{1}{2}\kappa+(2n-1)b}{\Delta^2+[\frac{1}{2}\kappa+(2n-1)b]^2} . \tag{7}$$

Introducing the ratio $q^n = W^n/W^{n-1}$ we obtain the continued-fraction expansion[21]

$$q^n = \frac{b_n}{1-a_{n+1}q^{n+1}} = \cfrac{b_n}{1-\cfrac{a_{n+1}b_{n+1}}{1-\cfrac{a_{n+2}b_{n+2}}{1-\cdots}}} , \tag{8a}$$

and can write the solution of Eq. (6) as

$$\langle w(t)\rangle = w^0 = \langle w(t)\rangle_{\rm DA}[1/(1-a_1q^1)] , \tag{8b}$$

$$w^n = \left(\prod_{j=1}^{n} q^j\right) w^0 . \tag{8c}$$

By application of Worpitzky's theorem[21] which states that continued fraction (8b) converges uniformly over any domain in which $|a_nb_n| \le \frac{1}{4}$ for $n=1,2,\ldots$, we find that the continued-fraction expansion for the averaged density-matrix elements converges for arbitrary Ω, κ, b, and Δ.

III. DISCUSSION

Before investigating the general exact solution (8), let us discuss the two limiting cases of large and zero-bandwidth fields. For $\Omega \ll b$ the continued fraction may be truncated in the first step neglecting terms of the order of $(\Omega/b)^4$, so that $\langle w(t)\rangle = \langle w(t)\rangle_{\rm DA}$, i.e., we recover the decorrelation result. For zero bandwidth fields ($b=0$), the continued fraction (8b) simplifies to

$$\langle w(t)\rangle = \langle w(t)\rangle_{\rm DA}\,(1+\kappa/A_1)\,e^{\kappa/A_1}E_1(\kappa/A_1), \tag{9}$$

with E_1 denoting the exponential integral.[20] This result is expected since for zero-bandwidth fields the statistical averaging reduces to an average of the population inversion in the monochromatic coherent field with respect to the P-distribution function[17]

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

of the CF[14]:

$$\langle w(t)\rangle^{b=0} = \int d^2\epsilon\, P(\epsilon,\epsilon^*)\frac{-\kappa}{\kappa+4\mu^2|\epsilon|^2[\frac{1}{2}\kappa/(\Delta^2+\frac{1}{4}\kappa^2)]}, \tag{10}$$

which may be shown to be identical with (9).

In general, the continued fraction for the population inversion must be calculated numerically. Results of these calculations are presented in Figs. 1–3. In these figures the excited state population $\langle\rho_{11}(t)\rangle$ is compared with the corresponding population $\langle\rho_{11}(t)\rangle_{\rm DA}$ in the DA or, equivalently, the population according to the phase diffusion model.[3] The dependence of the ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ on Ω for $\Delta=0$ and $b=0$, 0.2, 0.5, 1, 2, and 5κ is given in Fig. 1. As expected, this ratio is close to 1 in the weak field—where the DA is valid for any field. However, with increasing intensity, the ratio drops off very rapidly until a minimum is reached. This minimum at $\Omega\approx\kappa$ is most pronounced for $b=0$, where $\langle\rho_{11}\rangle = 0.81\,\langle\rho_{11}\rangle_{\rm DA}$. As b increases, it becomes shallower and shifts to larger values of Ω. Finally, for high intensities ($\Omega\gg\kappa, b$), the ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ tends again to one, since both $\langle\rho_{11}\rangle$ and $\langle\rho_{11}\rangle_{\rm DA}$ saturate at $\frac{1}{2}$. Thus, the population in the excited state is always less for an ideal CF than for a phase diffusing field.

Figure 2 shows $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ as a function of the bandwidth b for $\Omega=0.2$, 0.5, 1, 2, 5, and 10κ and $\Delta=0$. For small bandwidth, we have the drastic lowering of the ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ as a function of Ω which we already noted in Fig. 1. While for $\Omega\approx\kappa$ a small nonzero bandwidth immediately raises $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ close to 1, for $\Omega\gg\kappa$ a minimum in the bandwidth dependence appears near $b\approx\kappa$. With increasing Ω, this minimum flattens and shifts to larger values of b. In the large bandwidth limit $b\gg\Omega,\kappa$ all curves in Fig. 2 tend asymptotically to one in agreement with the DA.

The dependence on the detuning Δ is given in Fig. 3 for $b=0$, $\Omega=0.2$, 1, and 5κ and $b=\kappa$, $\Omega=0.2$, 1, and 5κ. Again we find $\langle\rho_{11}\rangle\approx\langle\rho_{11}\rangle_{\rm DA}$ for small Rabi frequencies and, therefore, the resonance curve for $\langle\rho_{11}\rangle$ has Lorentzian line shape. For $\Omega\approx\kappa$ the ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ shows a deep minimum near $\Delta=0$ which is most pronounced for $b=0$. Therefore, the on-resonance maximum of the dispersion curve for $\Omega\approx\kappa$ increases more slowly with Ω than for phase diffusing light. At high intensity $\Omega\gg\kappa$, when both resonance peaks of $\langle\rho_{11}\rangle$ and $\langle\rho_{11}\rangle_{\rm DA}$ saturate and their ratio is close to one, $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ drops sharply off-resonance until a minimum is reached. This minimum is again most pronounced for $b=0$. An increasing band-

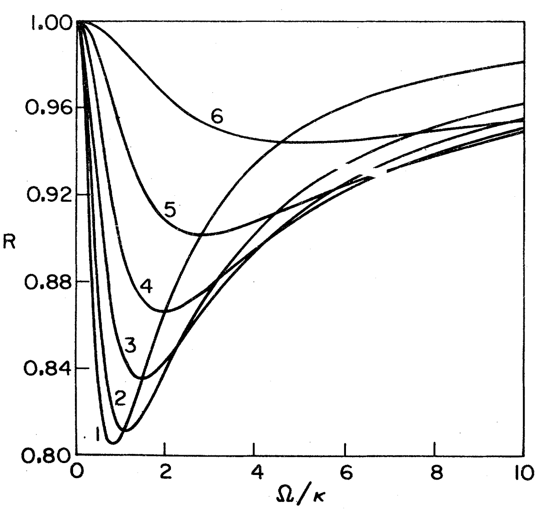

FIG. 1. Ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ is shown as function of Ω for $\Delta=0$. The curve index running from 1 to 6 denotes results for $b=0$, 0.2, 0.5, 1, 2, and 5.

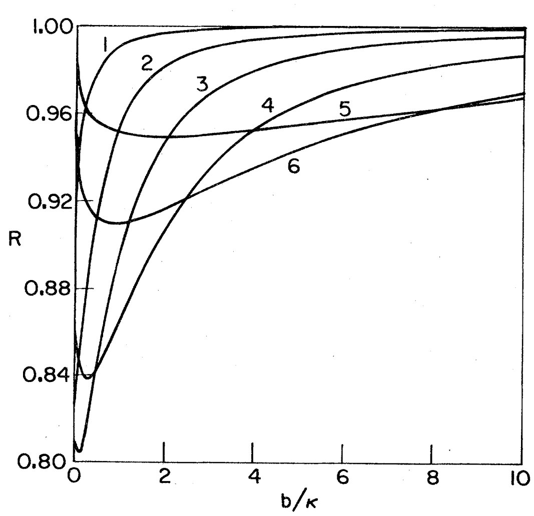

FIG. 2. Ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{\rm DA}$ is shown as function of b for $\Delta=0$. The curve index running from 1 to 6 corresponds to $\Omega=0.2$, 0.5, 1, 2, 5, and 10.

width tends to smooth this behavior. For large detuning $\Delta \gg \Omega$, when perturbation theory becomes valid, the ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{DA}$ approaches 1 again.

From the above discussion we infer that the DA is accurate to 20% or better in predicting the stationary populations of the TLA. This good agreement is a peculiarity of the TLA and may be misleading in some respects. Every approximate solution for the TLA with saturation included—such as the one obtained by the DA—predicts $\langle\rho_{11}\rangle \to \frac{1}{2}$ so that $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{DA} \to 1$ in the high-intensity limit. The behavior $\langle\rho_{11}\rangle \to \langle\rho_{11}\rangle_{DA}$ cannot be expected for an atomic system in which many levels are populated in the saturation regime and share the populations in a more complicated way. For the TLA, the disagreement of the exact solution and the DA becomes apparent when instead of $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{DA}$ we consider $\langle w\rangle/\langle w\rangle_{DA}$, the ratio of the population inversion, since both $\langle w\rangle$ and $\langle w\rangle_{DA}$ approach zero for high intensities. In particular, for $b=\Delta=0$ and $\Omega \gg \kappa$, from Eq. (9) we find

$$\langle w\rangle/\langle w\rangle_{DA} = \ln(2\Omega^2/\kappa^2) - 0.577\ ,$$

which increases with intensity. A similar behavior is found for finite values of b.

It will take considerably more theoretical efforts to get a complete understanding of resonant atomic processes in intense nonmonochromatic CF. It seems that the method employed in this paper is directly applicable to more complicated problems such as resonance fluorescence under a chaotic field, ac Stark splitting in double resonance[11] and, finally, multiphoton ionization,[14] where a full time-dependent treatment is often required.

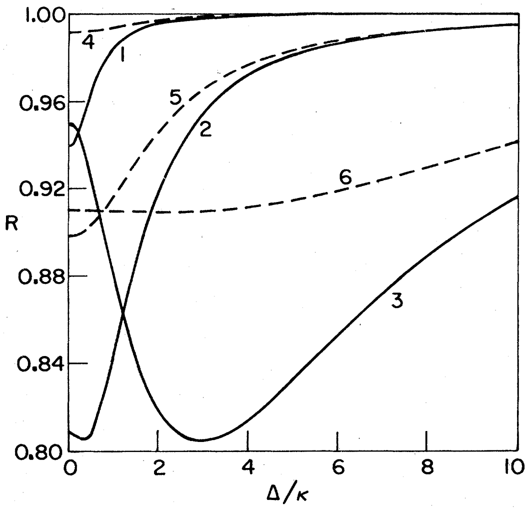

FIG. 3. Ratio $\langle\rho_{11}\rangle/\langle\rho_{11}\rangle_{DA}$ is shown as function of Δ. The curve index running from 1 to 6 corresponds to $b=0$, $\Omega=0.2$, 1, 5, and $b=1$, $\Omega=0.2$, 1, and 5.

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