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Light statistical dependence of saturated two-photon transitions

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Abstract. The light statistical dependence of stationary saturated two-photon transitions is discussed. While at low intensities, in comparison with light having a stable amplitude, Gaussian amplitude fluctuations of the exciting radiation are more effective in populating the excited resonant state, light with amplitude fluctuations is less effective in saturating a two-photon transition. The amplitude fluctuations are shown to wash out the resonance curves. While at low intensities the effective Stark shift (i.e. the maximum of the dispersion curve) is larger by a factor of three in light with Gaussian amplitude fluctuations compared with radiation of constant amplitude, this enhancement factor becomes intensity dependent with increasing intensity.

1. Introduction

The description of resonance phenomena in laser–atom interactions with high-power lasers has necessitated a theory capable of describing laser temporal coherence effects (laser light statistics) in these processes (Agostini *et al* 1978, Smith and Hogan 1979). The light statistical dependence of weak-field one-photon processes is completely determined by the mean intensity and spectrum (i.e. the Fourier transform of the first-order correlation function) of the exciting radiation. Similarly, laser temporal coherence effects of non-resonant N-photon transitions in weak fields are characterised completely by the Nth-order electric field correlation function (Lambropoulos 1976). A typical feature of saturated resonant atomic processes is their dependence on the infinite sequence of field correlation functions (Glauber 1965), corresponding to the infinite sequence of (virtual) up and down transitions of the electron between the resonant levels, which are responsible for the highly non-linear behaviour of the saturation of an atomic transition (Georges *et al* 1979). The summation of all the resonant terms in the perturbation series is a formidable mathematical problem. No general solutions are available; instead, this problem must be solved separately for each model of stochastic behaviour of the exciting radiation. Two types of fluctuation models have received particular interest in this context: the phase diffusion model (PDM) corresponding to a single-mode laser with a diffusing phase and the chaotic field (CF) model describing light from a multimode laser with a large number of uncorrelated modes (Glauber 1965, Zoller 1980). For the PDM the averaging over the fluctuations turns out to be rather simple as the atom–field averages can be decorrelated (see, for example, Agarwal 1978 and references quoted therein). For the CF such a decorrelation is impossible even in an approximate sense (Georges *et al* 1979); instead, non-perturbative methods such as the diagrammatic summation method (Elyutin 1977, Georges and Lambropoulos 1979) or Fokker–Planck eigenfunction techniques had to be developed (Zoller 1979a, b).

The application of these non-perturbative methods for the CF has until now been mainly limited to saturated one-photon transitions (Georges and Lambropoulos 1979, Zoller 1979a, b). In particular, the excited-state population of a strongly driven two-level system has been studied for the PDM and the CF with the result that the CF is less effective in saturating an atomic resonance compared with a PDM of the same mean intensity and bandwidth (Georges and Lambropoulos 1979). Even more dramatic changes have been predicted for the spectrum of resonance fluorescence and double optical resonance in agreement with experiment (Georges and Lambropoulos 1979, Zoller 1979a). In some aspects the saturated two-photon transition might be a more promising candidate to check these theoretical predictions regarding the difference in the saturation behaviour in the CF and the PDM. By saturated two-photon transitions we mean the process where an atomic ground state is coupled to the excited state by the infinite sequence of (virtual) two-photon emissions and absorptions. Since the Rabi frequency for the two-photon resonance is proportional to the light intensity (and not proportional to the electric field amplitude as for one-photon transitions) the saturation of a two-photon resonance shows a higher degree of non-linearity. Thus, one would expect a greater difference between the effects of the CF and the PDM. In addition, Stark shifts are important in two-photon transitions because they are usually of the same order of magnitude as the Rabi frequency (Agostini *et al* 1978). If intensity fluctuations are present, the Stark shift will be fluctuating and will tend to wash out the resonance curve. The maximum of the dispersion curve will, therefore, in general not appear at the frequency expected from the mean value of the Stark shift, but will be shifted by a different amount. This offers the possibility of studying the light statistical dependence of Stark shifts in a CF.

Recently, Georges and Lambropoulos (1979) have studied the stationary populations in a saturated two-level system coupled by a two-photon transition. They found that for low intensities the excited-state population in the CF was enhanced by a factor of 2! compared with the coherent field, while with increasing saturation the ratio of the population in the CF to the one in the coherent field shows a minimum with a value less than one and finally approaches one for high intensities. Their treatment, however, neglects Stark shifts and the bandwidth of the radiation. Saturated two-photon resonances have also been discussed in a recent paper on two-photon resonant three-photon ionisation within the Fokker–Planck eigenfunction formalism (Zoller and Lambropoulos 1980). Stark shifts and the finite bandwidth have been included in this treatment. The intrinsic time dependence of the multiphoton ionisation process, however allows, in view of computer time, the ionisation probability to be calculated with high accuracy only over a limited parameter range. It seems necessary, therefore, to complement the above investigations by calculations within a simple model which allows an explicit solution of arbitrary accuracy over a wide range of parameters.

In this paper we shall study such a model. We shall investigate the stationary excited-state populations in a two-photon coupled two-level system in a CF and a PDM. We shall discuss the intensity dependence of the ratio of the excited-state population for the two model fields, the effective Stark shifts and the resonance curves.

2. The model

We consider the interaction of a two-level atom with an electromagnetic light wave with an electric field vector of the form $\boldsymbol{E} = \boldsymbol{e}\mathscr{E}(t)\,\mathrm{e}^{-\mathrm{i}\omega t} + \mathrm{cc}$. The two atomic levels $|0\rangle$ and $|1\rangle$ are assumed to be coupled resonantly by two-photon transitions via the non-resonant states of the atom. $\mathscr{E}(t)$ is the stochastic amplitude of the electric field whose statistical properties will be specified below. ω is the mean frequency of the radiation. The optical Bloch equations of this two-level system are given by (Kimble and Mandel 1977)

$$\left[\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}+\begin{pmatrix} (\Delta-\delta\omega) & 0 & -\mu\mathscr{E}(t)^2 & \mu\mathscr{E}(t)^2 \\ 0 & -(\Delta-\delta\omega)+\frac{1}{2}\mathrm{i}\kappa & \mu\mathscr{E}^*(t)^2 & -\mu\mathscr{E}^*(t)^2 \\ -\mu\mathscr{E}^*(t)^2 & \mu\mathscr{E}(t)^2 & \mathrm{i}\kappa & 0 \\ \mu\mathscr{E}^*(t)^2 & -\mu\mathscr{E}(t)^2 & -\mathrm{i}\kappa & 0 \end{pmatrix}\right]\begin{pmatrix}\rho_{10}(t)\\ \rho_{01}(t)\\ \rho_{11}(t)\\ \rho_{00}(t)\end{pmatrix}=0 \tag{1}$$

with $\rho_{ij}(t)$ $(i, j = 0, 1)$ being the slowly varying density matrix elements. $\Delta = 2\omega - \omega_{10}$ denotes the detuning from resonance, κ is the spontaneous decay rate of level $|1\rangle$, μ represents the effective two-photon coupling matrix element $\mu = \Sigma_i(\mu_{0i}\cdot\boldsymbol{e})(\mu_{i1}\cdot\boldsymbol{e})/(\omega_{i0}-\omega)$, while $\delta\omega$ is the quadratic Stark shift of the transition $|0\rangle\to|1\rangle$.

Because of the stochastic nature of $\mathscr{E}(t)$, the optical Bloch equations (1) are a system of stochastic differential equations. It is our purpose to solve them for the averaged excited-state population $\langle\rho_{11}(t)\rangle$, with $\langle\ \rangle$ denoting the averaging over the fluctuations of $\mathscr{E}(t)$. In particular, we shall study below two different stochastic radiation models, the phase diffusion model (PDM) and the chaotic field (CF) model. The PDM describes a single-mode laser with a stable amplitude, but a slowly diffusing phase $\phi(t)$, obeying the Langevin equation $\dot\phi(t) = F_\phi(t)$. $F_\phi(t)$ is a random Gaussian force with $\langle F_\phi(t)F_\phi(t')\rangle = 2b\delta(t-t')$. The spectrum of the light in the PDM is Lorentzian with bandwidth b. Higher order correlation functions can be shown to have the factorisation property (Georges and Lambropoulos 1979)

$$\langle\mathscr{E}^{*N}(t_1)\mathscr{E}^{M}(t_2)\ldots\mathscr{E}^{*N}(t_{2n-1})\mathscr{E}^{N}(t_{2n})\rangle=\prod_{i=1}^{n}\langle\mathscr{E}^{*N}(t_{2i-1})\mathscr{E}^{N}(t_{2i})\rangle \tag{2}$$

for $t_1\geqslant t_2\geqslant\ldots\geqslant t_{2n}$. Two-time correlation functions are given by

$$\langle\mathscr{E}^{*N}(t_1)\mathscr{E}^{N}(t_2)\rangle=|\mathscr{E}_0|^{2N}\exp(-N^2b|t_1-t_2|). \tag{3}$$

The CF is a field exhibiting Gaussian amplitude fluctuations. Typically the higher order correlation functions of a CF fulfill (Glauber 1965)

$$\langle\mathscr{E}^*(t_1)\mathscr{E}^*(t_2)\ldots\mathscr{E}(t_{2n-1})\mathscr{E}(t_{2n})\rangle=\sum_P\prod_{i=1}^{n}\langle\mathscr{E}^*(t_i)\mathscr{E}(t_{P(i+n)})\rangle \tag{4}$$

where P denotes permutations. In this case the two-time correlation functions are given by

$$\langle\mathscr{E}^{*N}(t_1)\mathscr{E}^{N}(t_2)\rangle=N!\langle|\mathscr{E}|^2\rangle\exp(-Nb|t_1-t_2|). \tag{5}$$

Examples of a CF are light from a thermal source and radiation from a (pulsed) multimode laser with a large number of uncorrelated modes. A Markovian model for a CF can be described by the Langevin equations (Zoller 1979a, b)

$$\dot{\mathscr{E}}(t)=-b\mathscr{E}(t)+F_{\mathscr{E}}(t)\qquad\dot{\mathscr{E}}^*(t)=-b\mathscr{E}^*(t)+F_{\mathscr{E}^*}(t) \tag{6}$$

with $\langle F_{\mathscr{E}}(t)F_{\mathscr{E}^*}(t')\rangle = 2\langle|\mathscr{E}|^2\rangle b\delta(t-t')$. The spectrum of the CF described by (5) is a Lorentzian with bandwidth b.

3. Saturation of a two-photon transition

We are looking for solutions of the stochastic optical Bloch equations (1), averaged over the fluctuations of the electric field amplitude $\mathscr{E}(t)$. In particular, we are interested in the stationary population of state $|1\rangle$ after all transient effects have died out.

For the PDM the averaging over the field fluctuations is particularly simple (Avan and Cohen-Tannoudji 1977, Agarwal 1976, Eberly 1976, Kimble and Mandel 1977, Georges and Lambropoulos 1978, Wódkiewicz 1979). Due to the factorisation property (2) of the electric field correlation functions, the atomic populations can be rigorously decorrelated from the field variables at an appropriate stage of the calculation. It can be shown (Agarwal 1976, Wódkiewicz 1979) that the averaging replaces the transverse decay constant $\kappa/2$ in the optical Bloch equations (1) by $\kappa/2+4b$. Note that the additional factor of $4b$ stems from the relaxation constant of the second-order field correlation function (3) with $N=2$. Thus we find for the averaged excited-state population for the PDM

$$\langle\rho_{11}\rangle_{\mathrm{PD}} = W^{\mathrm{PD}}/(\kappa+2W^{\mathrm{PD}}) \tag{7}$$

with

$$W^{\mathrm{PD}} = \frac{\frac{1}{2}\Omega^2(4b+\kappa/2)}{(\Delta-\delta\omega)^2+(4b+\kappa/2)^2} \tag{8}$$

where $\Omega = 2\mu|\mathscr{E}(t)|^2$. W^{PD} may be interpreted as the transition rate $|0\rangle-|1\rangle$.

For the CF, on the other hand, the averaging over the fluctuations is much more complicated because a decorrelation of the atom–field variables is possible only for large bandwidth fields b, $\kappa \gg \langle\Omega\rangle$, $\langle\delta\omega\rangle$, but becomes inadequate in the saturation regime. In the particular case of zero bandwidth field ($b=0$), $\langle\rho_{11}(t)\rangle$ may be found by averaging the excited-state population in the coherent field, namely

$$\rho_{11}(\mathscr{E},\mathscr{E}^*) = \tfrac{1}{2}\Omega^2\frac{\kappa/2}{(\Delta-\delta\omega)^2+(\kappa/2)^2}\left(\kappa+2\tfrac{1}{2}\Omega^2\frac{\kappa/2}{(\Delta-\delta\omega)^2+(\kappa/2)^2}\right)^{-1} \tag{9}$$

over the Gaussian amplitude distribution $P_S(\mathscr{E},\mathscr{E}^*) = [1/\pi\langle|\mathscr{E}|^2\rangle]\exp-(|\mathscr{E}|^2/\langle|\mathscr{E}|^2\rangle)$ of the CF (Glauber 1965):

$$\begin{aligned}
\langle\rho_{11}\rangle &\overset{b=0}{=} \int \mathrm{d}^2\mathscr{E} P_S(\mathscr{E},\mathscr{E}^*)\rho_{11}(\mathscr{E},\mathscr{E}^*) = \frac{1}{4}\frac{\langle\Omega\rangle^2}{\frac{1}{2}\langle\Omega\rangle^2+\langle\delta\omega\rangle^2} + \mathrm{Re}\,(T\mathrm{e}^{-x}E_1(x)) \\
x &= \Delta\frac{\langle\delta\omega\rangle}{\frac{1}{2}\langle\Omega\rangle^2+\langle\delta\omega\rangle^2} + \mathrm{i}\frac{\langle\Omega\rangle}{\frac{1}{2}\langle\Omega\rangle^2+\langle\delta\omega\rangle^2}[\tfrac{1}{2}(\Delta^2+\tfrac{1}{4}\kappa^2)+\tfrac{1}{4}\kappa^2(\langle\delta\omega\rangle/\langle\Omega\rangle)^2]^{1/2} \\
R &= -\frac{1}{2}\frac{\Delta\langle\Omega\rangle^2\langle\delta\omega\rangle^2}{(\frac{1}{2}\langle\Omega\rangle^2+\langle\delta\omega\rangle^2)^2} - \tfrac{1}{4}\mathrm{i}\langle\Omega\rangle\frac{\frac{1}{2}\langle\Omega\rangle^2(\Delta^2+\kappa^2/4)+\langle\delta\omega\rangle^2(\kappa^2/4-\Delta^2)}{(\frac{1}{2}\langle\Omega\rangle^2+\langle\delta\omega\rangle^2)^2[\frac{1}{2}(\Delta^2+\kappa^2/4)+\kappa^2/4(\langle\delta\omega\rangle/\langle\Omega\rangle)^2]^{1/2}}.
\end{aligned} \tag{10}$$

For arbitrary bandwidth it can be shown that the stochastic optical Bloch equations (1) may be transformed to an infinite system of differential equations for the averages

$\rho_{ij}^{\alpha n}(t)$ (Zoller 1979b)

$$\left(\frac{\mathrm{d}}{\mathrm{d}t}+\kappa+2nb\right)\rho_{11}^{0n}(t)=\tfrac{1}{2}\mathrm{i}\langle\Omega\rangle[\sqrt{(n+1)(n+2)}(\rho_{01}^{2n}(t)-\rho_{10}^{-2n}(t))$$
$$-2\sqrt{n(n+1)}(\rho_{01}^{2n-1}(t)-\rho_{10}^{-2n-1}(t))+\sqrt{n(n-1)}(\rho_{01}^{2n-2}(t)-\rho_{10}^{-2n-2}(t))]$$
$$\left(\frac{\mathrm{d}}{\mathrm{d}t}+\mathrm{i}\Delta-\mathrm{i}\langle\delta\omega\rangle(2n+3)+\tfrac{1}{2}\kappa+2b(n+1)\right)\rho_{01}^{2n}(t)=\tfrac{1}{2}\mathrm{i}\langle\Omega\rangle\sqrt{(n+1)(n+2)}(W^{n}(t)$$
$$-2W^{n+1}(t)+W^{n+2}(t))-\mathrm{i}\langle\delta\omega\rangle(\sqrt{(n+1)(n+3)}\rho_{01}^{2n+1}(t)$$
$$+\sqrt{n(n+2)}\rho_{01}^{2n-1}(t)) \qquad (11)$$

where $W^{n}(t)=\rho_{11}^{0n}(t)-\rho_{00}^{0n}(t)$ and

$$\rho_{ij}^{\alpha n}(t)=(n!/(n+|\alpha|)!)^{1/2}\langle|\mathscr{E}(t)|^{|\alpha|}(\mathscr{E}(t)/\mathscr{E}^{*}(t))^{\alpha/2}L_{n}^{|\alpha|}(|\mathscr{E}(t)|^{2}/\langle|\mathscr{E}|\rangle^{2})\rho_{ij}(t)\rangle/\langle|\mathscr{E}|^{2}\rangle^{|\alpha|/2} \qquad (12)$$

with $L_n^{|\alpha|}$ being Laguerre polynomials. Note that the average excited-state population is given by $\langle\rho_{11}(t)\rangle_{\mathrm{CF}}=\rho_{11}^{00}(t)$. In the stationary limit $\dot{\rho}_{ij}^{\alpha n}=0$ we can derive from (11) a five-term recursion relation

$$S^{n}[D_{n}^{2}/A_{n}+A_{n}+(n+1)(n+2)(B_{n}+4B_{n+1}+B_{n+2})+(\langle\delta\omega\rangle/\langle\Omega\rangle)^{2}((n+1)(n+3)/A_{n+1}$$
$$+n(n+2)/A_{n-1})]+S^{n+1}\sqrt{(n+1)(n+3)}$$
$$\times[D_{n}\langle\delta\omega\rangle/(A_{n}\langle\Omega\rangle)-2(n+2)(B_{n+1}+B_{n+2})+\langle\delta\omega\rangle D_{n+1}/(\langle\Omega\rangle A_{n+1})]$$
$$+S^{n-1}\sqrt{n(n+2)}[D_{n}\langle\delta\omega\rangle/(A_{n}\langle\Omega\rangle)-2(n+1)(B_{n}+B_{n+1})$$
$$+\langle\delta\omega\rangle D_{n-1}/(\langle\Omega\rangle A_{n-1})]$$
$$+S^{n+2}\sqrt{(n+1)(n+2)(n+3)(n+4)}[B_{n+2}+(\langle\delta\omega\rangle/\langle\Omega\rangle)^{2}/A_{n+1}]$$
$$+S^{n-2}\sqrt{(n-1)n(n+1)(n+2)}[B_{n}+(\langle\delta\omega\rangle/\langle\Omega\rangle)^{2}/A_{n-1}]=-1/\sqrt{2}\delta_{n0} \qquad (13)$$

with

$$S^{n}=\operatorname{Im}\{\rho_{01}^{2n}\}$$
$$A_{n}=(\kappa/2+2b(n+1))/\langle\Omega\rangle$$
$$B_{n}=\langle\Omega\rangle/(\kappa+2bn)$$

and

$$D_{n}=(\Delta-\langle\delta\omega\rangle(2n+3))/\langle\Omega\rangle.$$

The average $\langle\rho_{11}\rangle_{\mathrm{CF}}$ is then given by

$$\langle\rho_{11}\rangle_{\mathrm{CF}}=-\sqrt{2}\langle\Omega\rangle S^{0}/\kappa. \qquad (14)$$

According to (13) the coupling of S^0 to all higher order S^n indicates the influence of the higher order statistics on the saturation behaviour.

In the weak-field limit $\langle\Omega\rangle$, $\langle\delta\omega\rangle\ll\kappa$, b, neglecting the coupling of S^0 to the higher order averages, we again find a result of the form (7) with W^{PD} replaced by the transition rate

$$W^{\mathrm{CF}}=2!\tfrac{1}{2}\langle\Omega\rangle^{2}\frac{\kappa/2+2b}{(\Delta-3\langle\delta\omega\rangle)^{2}+(\kappa/2+2b)^{2}}. \qquad (15)$$

This result is also obtained in the decorrelation approximation. Note that W^{CF} differs from W^{PD} in the overall factor of 2! and that the different factors by which the bandwidth is multiplied reflect the differences in the second-order correlation functions in the PDM and the CF. Furthermore, the Stark shift is enhanced by a factor of three in the CF as compared with the PDM.

For high intensities, we have solved the system of equations (13) by numerical methods. The results of these calculations will be discussed in the next section.

4. Discussion

Figure 1 shows the intensity dependence of the ratio of the populations in the excited state for the case of a CF and radiation obeying the PDM for $b = 0, 0{\cdot}5\kappa, 1\kappa, 2\kappa, 10\kappa$, $\Delta = 0$, $\langle\delta\omega\rangle = 0$. At low intensities ($W \ll \kappa$) this ratio is approximately given by the ratios of the induced bound–bound rates W^{CF}/W^{PD}. For $b = 0$ we therefore have $\langle\rho_{11}\rangle_{CF}/\langle\rho_{11}\rangle_{PD} \rightarrow 2!$. In the limit of large bandwidth fields this ratio goes to $2! \times 2$ with the factor of 2 stemming from the additional factor multiplying the bandwidth in the PDM. Increasing the intensity results in the lowering of the ratio $\langle\rho_{11}\rangle_{CF}/\langle\rho_{11}\rangle_{PD}$. For small bandwidth fields we may even find $\langle\rho_{11}\rangle_{CF}$ to be smaller than $\langle\rho_{11}\rangle_{PD}$ (Georges and Lambropoulos 1979). This minimum in the intensity dependence, however, disappears for large values of b. In the high-intensity limit $\langle\rho_{11}\rangle_{CF}/\langle\rho_{11}\rangle_{PD}$ approaches the value of one as the population of a strongly driven two-level system becomes one half, irrespective of the fluctuations in the exciting radiation field.

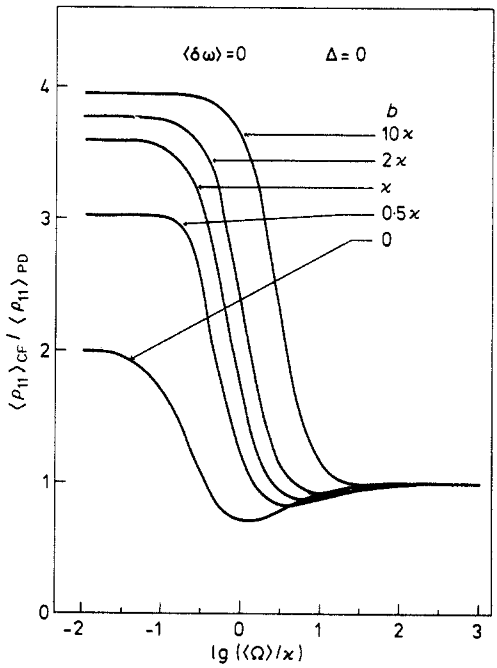

Figure 1. The ratio of the populations of the upper level for a CF and the PDM are plotted as a function of the mean Rabi frequency $\langle\Omega\rangle$ for various values of the laser bandwidth b.

Figure 2 compares the resonance curves for a CF and the PDM for different values of the mean Stark shifts. Note that for $b = 0$ the maximum of the dispersion curve is smaller in a CF than for the PDM, while the resonance curve in a CF has higher wings due to the off-resonance 2! enhancement. Whereas in a field with stable amplitude the resonance curves are simply shifted with increasing $\delta\omega$, the amplitude fluctuations of a CF wash out the resonance and cause an asymmetry. Also note that because of the correlation between the atomic transition and the Stark shift, the effective Stark shift $\delta\omega_{\text{eff}}$ (i.e. the maximum of the resonance curve) is different from the mean Stark shift in a CF, contrary to the case of the PDM.

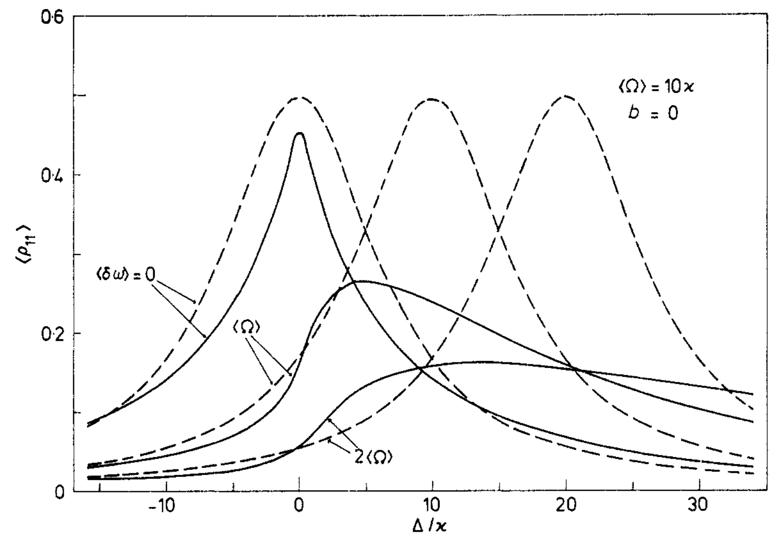

Figure 2. Resonance curves for a CF (full curves) and the PDM (broken curves) for certain values of the mean Stark shift $\langle\delta\omega\rangle$.

Figure 3 shows the dependence of the ratio of the effective Stark shift $\delta\omega_{\text{eff}}$ to the mean Stark shift $\langle\delta\omega\rangle$ on the mean Rabi frequency for certain values of the laser bandwidth b. In this figure the mean Stark shift is assumed to be equal to the mean Rabi frequency. Note that the maximum of the dispersion curve is not shifted by the mean value of the Stark shift $\langle\delta\omega\rangle$ since in the presence of intensity fluctuations the Stark shift $\delta\omega$ enters the intensity dependence of the excitation probability in a non-linear way (compare equations (9) and (10) and Agostini *et al* (1978)). For low intensities (i.e. $\langle\Omega\rangle$, $\langle\delta\omega\rangle \ll \kappa, b$) this ratio approaches three. This value $3 = 3!/2!$ arises because the Stark shift—being a third-order correction to the lowest second-order perturbation calculation of W^{CF}—is enhanced by the intensity fluctuations; with increasing intensity this ratio becomes even less than one. Note that at very high intensities the effective Stark shift is no longer well defined since the resonance curves become increasingly washed out by the intensity fluctuations.

If the two-level system loses population from the upper state by a weak-field induced one-photon process (for example, ionisation to the continuum), this transition rate will be proportional to $\langle|\mathscr{E}(t)|^2\rho_{11}(t)\rangle$. Including ionisation perturbatively in the two-level system is, of course, possible only for intensities for which the ionisation rate is smaller than the internal relaxation rates (spontaneous decay or similar relaxation

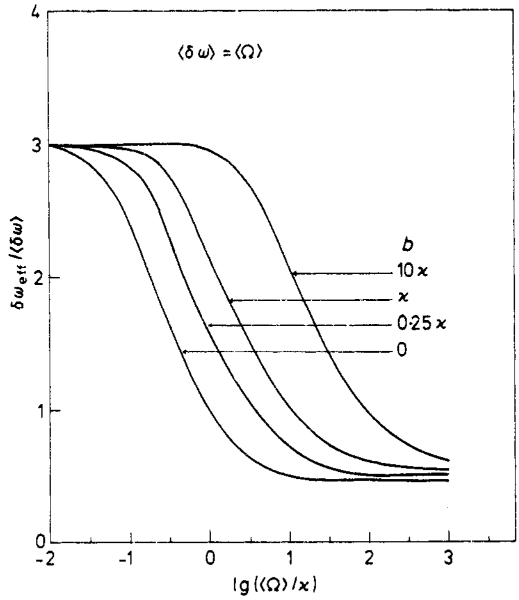

Figure 3. The ratio of the effective Stark shift $\delta\omega_{\rm eff}$ and the mean Stark shift $\langle\delta\omega\rangle$ as a function of the mean Rabi frequency $\langle\Omega\rangle$ for various values of the laser bandwidth b.

mechanisms). Although for many experiments this assumption is somewhat unrealistic, it is nonetheless interesting to compare the ratio of the ionisation probabilities per unit time $\langle|\mathscr{E}(t)|^2\rho_{11}(t)\rangle_{\rm CF}/\langle|\mathscr{E}(t)|^2\rho_{11}(t)\rangle_{\rm FD}$ in a CF and a phase diffusing field within our model. A time-dependent treatment of two-photon resonant three-photon ionisation has been given by Zoller and Lambropoulos (1980). This investigation was incomplete, however, in so far that only the first few terms in the expansion (11) of the solution of the equations (10) were taken into account. Figure 4 shows the ratio $\langle|\mathscr{E}|^2\rho_{11}\rangle_{\rm CF}/\langle|\mathscr{E}|^2\rho_{11}\rangle_{\rm PD}$ for $b = 0, 0{\cdot}5\kappa, 1\kappa, 2\kappa, 10\kappa$ with $\Delta = 0, \langle\delta\omega\rangle = 0$ as a function of the mean intensity. For low intensities we see that this ratio approaches 3! for small bandwidth fields (Lambropoulos 1976). A finite bandwidth brings in the different factors multiplying the bandwidth in the PDM and the CF and, therefore, increases this ratio. In the limit of large bandwidth, however, the bound–bound and bound–free steps become statistically independent i.e. $\langle|\mathscr{E}|^2\rho_{11}\rangle_{\rm CF} \simeq \langle|\mathscr{E}|^2\rangle\langle\rho_{11}\rangle_{\rm CF}$ and the ratio $\langle|\mathscr{E}|^2\rho_{11}\rangle_{\rm CF}/\langle|\mathscr{E}|^2\rho_{11}\rangle_{\rm PD}$ reduces to $\langle\rho_{11}\rangle_{\rm CF}/\langle\rho_{11}\rangle_{\rm PD} \simeq 4$. An increase of the intensity decreases these ratios.

In summary, we have investigated the effect of phase and amplitude fluctuations of a laser field on a resonant atomic two-photon transition. We have seen that the different higher order statistical properties of the radiation fields cause significant differences in the saturation behaviour of the atomic transition. Whereas for low intensities the statistical dependence is completely determined by the second-order correlation functions, the saturation behaviour exhibits a highly non-linear response to the fluctuations in the incident field. The ratio of the populations in the upper level for a CF and the PDM therefore, for low intensities, approaches the value of 2! for small bandwidth and $2!\times 2$ for large bandwidth fields. With increasing intensity we find a minimum even less than one for small bandwidth fields. In this case the PDM is more effective in populating the upper level, contrary to the low-intensity limit. For the resonance curves for a chaotic incident field higher wings have been found and

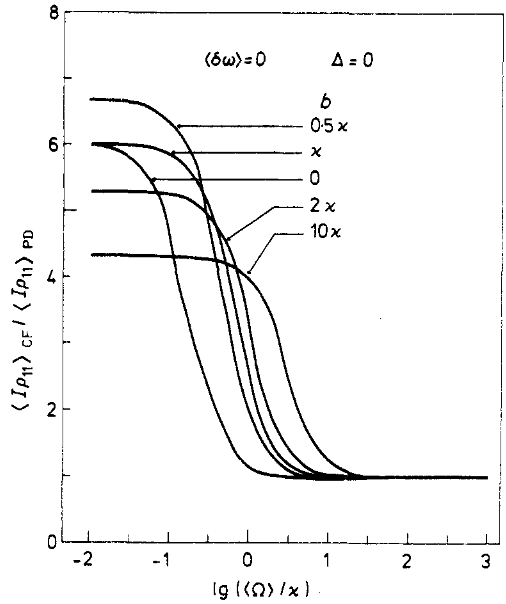

Figure 4. The ratio of the rates of a two-photon resonant three-photon ionisation for a CF and the PDM as a function of the mean Rabi frequency $\langle\Omega\rangle$.

moreover an asymmetry which is also brought about by the non-linear dependence of the Stark shifts on the amplitude fluctuations of the field. Because of the correlation between the atomic transition and the Stark shift in a CF, the effective Stark shift (i.e. the maximum of the resonance curve) is different from the mean Stark shift and depends on the intensity of the radiation. Within our model we also treated the effect of a two-photon resonant three-photon ionisation in a perturbative way. We found that in the small bandwidth case the atomic transition and the ionisation are correlated whereas for increasing bandwidth the excitation of the resonant intermediate state becomes statistically decoupled from the ionisation from the resonant state to the continuum.

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