Stark broadening may prevent resolution of neon satellite lines. This problem can be overcome by using seed ions with higher atomic number Z. The Stark width[14] of the L_α line is proportional to $Z^{-5.9}$, whereas the separation between the satellite lines[10] is proportional to $Z^{-2.8}$.

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Saturation and Stark Splitting of Resonant Transitions in Strong Chaotic Fields of Arbitrary Bandwidth

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We report the solution of the problem of saturation and ac Stark splitting of a resonant transition in a strong chaotic field of arbitrary bandwidth. We present results for double optical resonance and resonance fluorescence and compare them to those obtained for a phase-diffusion field.

The role of field fluctuations and of the associated bandwidth in the resonant interaction of intense radiation with matter has been an extremely active subject in the last three years or so. A number of interesting results[1-8] have been obtained under the assumption of a bandwidth due entirely to the fluctuations of the phase (phase diffusion) of the field, whose amplitude is assumed to be constant. The phase-diffusion model is, of course, adequate for the interpretation of experiments with well-stabilized cw lasers, notably those used in recent experiments on resonance fluorescence[9,10] under intense fields. There is, however, another class of experiments —such as multiphoton experiments,[11,12] for example—performed with considerably stronger, multimode, pulsed lasers whose amplitude undergoes substantial fluctuations, often comparable to, if not stronger than, those of a chaotic field. We have therefore a more general and far more significant problem: How does an intense, stochastically fluctuating field—with both amplitude and phase fluctuations—affect the saturation and the associated Stark splitting of a resonant transition? It also is a far more difficult problem that had thus far eluded solution, although some of its aspects have been investigated.[13-18] In this Letter, we report the solution of the problem for a chaotic field and present results on saturation, resonance fluorescence, and double optical resonance.

The essential mathematical problem, which can be formulated in more than one way, basically requires the solution of the equations of motion of

a two-level system (TLS) strongly coupled to a stochastically fluctuating field. Thus one has to solve a set of stochastic differential equations. For phase diffusion, the task is facilitated significantly because the atomic variables can at the appropriate stage of the calculation be decorrelated rigorously from the field variables. The decorrelation refers to the stochastic averaging over the fluctuating variables. As a result, the stochastic differential equations are reduced to differential equations obeyed by averaged atomic variables. But for amplitude fluctuations, the decorrelation is not valid and if it is used as an approximation one does not know the magnitude of the error, unless of course the solution of the complete problem is known. It is only in the weak-field limit that the decorrelation is valid for all fields.

We present here results from the solution of the problem for a chaotic field which, as is well known, undergoes Gaussian amplitude fluctuations. These results have been obtained with two parallel approaches.[19-21] In one of the approaches,[19] the chaotic field is assumed to be Markovian and is represented by its Fokker-Planck equation. As has been shown elsewhere,[19] if one is interested in certain one-time atom-field averages, the stochastic density-matrix equations can be reduced to an infinite set of differential equations for these averages. If, in addition, one considers the stationary limit of the averaged density matrix of the TLS, the equations reduce to an infinite system of linear algebraic equations which can be used to obtain solutions in terms of continued fractions. Let $\rho_{11}(t)$ and $\rho_{22}(t)$ be diagonal matrix elements (populations) of the density matrix of the TLS, where $|2\rangle$ and $|1\rangle$ are the upper and lower states, respectively, with energies $\hbar\omega_2$ and $\hbar\omega_1$. These matrix elements are, of course, coupled to $\rho_{12}(t)$. The stationary limit of the average (indicated by angular brackets) population difference $\langle n(t)\rangle \equiv \langle \rho_{22}(t)\rangle - \langle \rho_{11}(t)\rangle$ corresponds to $t \to \infty$ and can be written as a continued fraction involving the averaged Rabi frequency Ω, the detuning Δ from resonance, the field bandwidth b, and the spontaneous decay width κ_2 of the upper state. This continued fraction has been shown[19] to converge for all values of the above parameters. In general, it must be calculated numerically but the convergence is very rapid which enables one to obtain results for arbitrary field intensities and bandwidths.

In the second approach,[21] the chaotic field $\epsilon(t)$ is written as a complex Gaussian stochastic process described by the infinite sequence of its field-correlation functions. Although such a process is not necessarily Markovian, we do here assume a Markovian chaotic field with first-order correlation function $\langle \mathcal{E}^*(t_1)\mathcal{E}(t_2)\rangle = \mathcal{E}_0^2 \exp(-b|t_1 - t_2|)$, where $\mathcal{E}_0^2 \equiv \langle |\mathcal{E}(t)|^2\rangle$, and the resulting spectrum is obviously Lorentzian. The correlation functions of the chaotic field obey well-known relations.[22] Again using the density-matrix equations, an integral equation for $n(t)$ can be obtained. But in attempting to calculate $\langle n(t)\rangle$ one encounters correlations of the form $\langle \mathcal{E}^*(t_1)\mathcal{E}(t_2)n(t_2)\rangle$ in which the field and atom variables can be decorrelated only for a phase-diffusion field. To solve the problem for the chaotic field, we have used the correlation functions of the field to obtain a series expansion for $\langle \mathcal{E}^*(t_1)\mathcal{E}(t_2)n(t_2)\rangle$. When this is substituted into the integral equation for $\langle n(t)\rangle$, then after an iteration procedure whose details will be presented elsewhere,[21] we were able to obtain a series integral equation which can be written in a diagrammatic form. Taking the Laplace transform, one can then express $\langle n(t=\infty)\rangle$ in terms of a continued fraction equivalent to the one of the first approach.

Before presenting some of the results obtained with the above approaches, it is worth discussing an analytical result that illustrates much of the physics involved. For a chaotic field of zero bandwidth (physically corresponding to a bandwidth much smaller than the natural width of the atomic transition), the continued fraction simplifies considerably and $\langle n(\infty)\rangle$ can be written as

$$\langle n(\infty)\rangle^{\mathrm{CH}} = \langle n(\infty)\rangle^{\mathrm{PD}} (1 + 1/S) e^{1/S} E_1(1/S) \qquad (1)$$

where $S \equiv \frac{1}{2}\Omega^2/(\Delta^2 + \frac{1}{4}\kappa_2^2)$ is the usual saturation parameter of the TLS in a monochromatic field: $\Omega = 2\hbar^{-1}\mu_{12}\mathcal{E}_0$ is the average Rabi frequency, and E_1 is the exponential integral. Here CH indicates chaotic and PD phase diffusion. Note that $\langle n(\infty)\rangle^{\mathrm{PD}} = -(1+S)^{-1}$. From the properties of the exponential integral one finds that $\langle n(\infty)\rangle^{\mathrm{CH}}$ and $\langle n(\infty)\rangle^{\mathrm{PD}}$ are equal, to first order in S (weak field); for large S, $\langle n(\infty)\rangle^{\mathrm{CH}} \simeq -\ln S/S$ while $\langle n(\infty)\rangle^{\mathrm{PD}} \simeq -1/S$. As expected, in both cases $\langle n(\infty)\rangle$ tends to zero as $S \to \infty$, but it approaches zero more slowly for a chaotic than for a coherent field. This means that the chaotic field is less effective than the coherent field in saturating a one-photon transition. As we will see below, this turns out to be a basic feature of the chaotic field that persists for arbitrary bandwidth.

Let us now consider some of the results illustrating the effects of field fluctuations for chaotic

fields of arbitrary bandwidth. In Fig. 1 we plot the ratio $R \equiv \langle \rho_{22}(\infty) \rangle^{CH} / \langle \rho_{22}(\infty) \rangle^{PD}$ as a function of field intensity for various field bandwidths b. As we progress from curve 1 (corresponding to $b = 0$) to curve 6 (corresponding to $b = 5\kappa_2$) the bandwidth increases. For zero field, the ratio R is unity, but as the field increases, the ratio drops to a minimum value which depends on the bandwidth. Also the field strength at which the minimum occurs depends on the bandwidth. Curve 1 represents the analytical results of Eq. (1). The other curves have resulted from the numerical calculation of the continued fraction. Curve 2, for example, shows that for a bandwidth $b = 0.2\kappa_2$ the minimum occurs at a field strength for which $\Omega \simeq \kappa_2$. Clearly, the results illustrated by these curves prove an assertion made earlier: The chaotic field is always less effective than a phase-diffusion field in saturating a resonant transition. It also implies that the decorrelation approximation will be inaccurate—by a maximum of ~20% —if used for a chaotic field. The difference between chaotic and phase-diffusion fields decreases as the bandwidth increases becoming about 6% for $b = 5\kappa_2$. We note here in passing that we have found similar behavior also when $|1\rangle$ and $|2\rangle$ are coupled through an N-photon transition.[21, 23]

In Fig. 2, we present results for resonance fluorescence in a strong chaotic field. The dashed curves correspond to a phase-diffusion field and the solid curves to a chaotic field. In both cases, the calculation is for fields exactly on resonance and with bandwidth $b = \kappa_2$. Recall that for a monochromatic coherent field, the spectrum of the spontaneously emitted photons ω_k has the well-known triplet structure with the central peak at the frequency of the strong field, and the sidebands shifted by an amount equal to the Rabi frequency.[24] The overall structure is preserved in a phase-diffusion field of finite bandwidth. Under a chaotic field of the same bandwidth, the spectrum undergoes a dramatic change. The triplet structure is still there, but much less pronounced with only the central peak being clearly visible. The main reason for the near obliteration of the triplet structure is that the sidebands are smeared because the amplitude fluctuation causes fluctuation of the Rabi frequency. This suppression of the sidebands had been anticipated qualitatively by Avan and Cohen-Tannoudji,[7] but the triplet structure is not washed out completely as their argument had suggested. The central peak is affected somewhat by the amplitude fluctuations, but it broadens mainly because of the finite bandwidth. For a chaotic field of zero bandwidth, we have found that the central peak remains essentially unchanged while the triplet structure is smeared out. In that case, the splitting turns out to be equal to $\Omega/\sqrt{2}$ and not Ω. As is evident in Fig. 2, the on-resonance splitting for a chaotic field is smaller than that for a phase-diffusion

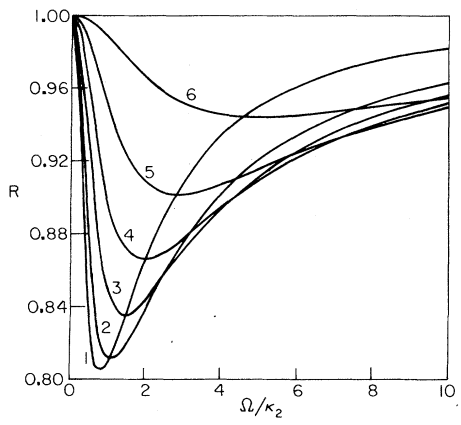

FIG. 1. Saturation of a two-level system under a strong stochastic field. The ratio $R = \langle \rho_{22}(\infty) \rangle^{CH} / \langle \rho_{22}(\infty) \rangle^{PD}$ is shown as a function of Ω/κ_2 for $\Delta = 0$. The curves numbered 1 to 6 correspond to field bandwidths $b = 0$, $0.2\kappa_2$, $0.5\kappa_2$, κ_2, $2\kappa_2$, and $5\kappa_2$.

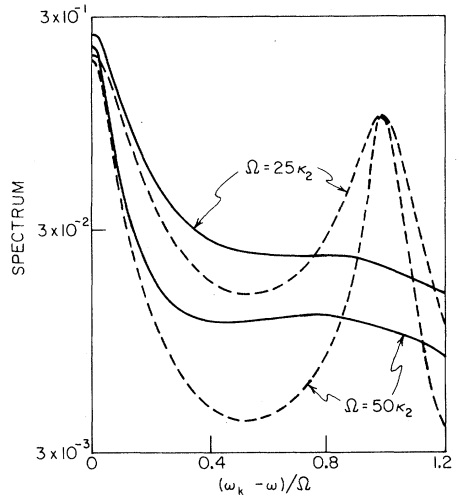

FIG. 2. The spectrum of resonance fluorescence under a strong stochastic field exactly on resonance ($\Delta = 0$) and of bandwidth $b = \kappa_2$. The dashed curves correspond to a PD field and the solid curves to a CH. The spectrum of the fluorescent photons ω_k is plotted as a function of $(\omega_k - \omega)/\Omega$, where ω is the atomic frequency and Ω the averaged Rabi frequency as defined in the text. Only half of the symmetric spectrum is shown.

field of the same bandwidth and intensity.

We turn finally to the effects of field fluctuations on Stark splitting in double optical resonance. The TLS is again coupled to a near-resonant strong stochastic field $[\mathcal{E}(t)e^{i\omega t}+\text{c.c.}]$ while a second weak stochastic field $[\mathcal{E}'(t)e^{i\omega' t}+\text{c.c.}]$ is nearly resonant with a second transition $|2\rangle \rightarrow |3\rangle$. In Fig. 3 we show results for two different intensities of strong chaotic fields of finite bandwidth. In the same figure, results corresponding to phase-diffusion fields of the same bandwidth and intensities are also shown. The most striking feature in Fig. 3 is the substantial broadening of the peaks caused by the amplitude fluctuations of the chaotic field. That this is not a result of the finite bandwidth alone becomes evident by noticing the difference from the corresponding curve for the phase-diffusion field. Our calculation corresponds to an experiment on Stark splitting in three-photon ionization that we have analyzed in a recent paper[12] using a phase-diffusion model. That analysis had left unanswered the question of the origin of the substantial broadening (beyond the laser bandwidth and ionization widths) that was evident in the experimental data. Our present results, as illustrated in Fig. 3, provide the answer. The laser field of the experiment did have amplitude fluctuations, although it may not have been chaotic. It must be stressed here that this additional broadening is present even for a chaotic field of zero bandwidth since it is an effect basically related to intensity fluctuations. We have also obtained a number of other new results especially for off-resonant excitation which will be presented elsewhere.

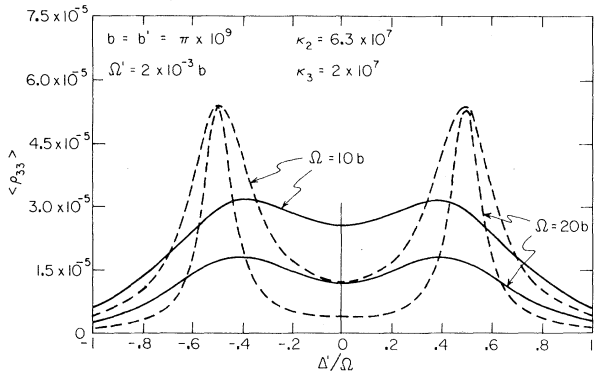

FIG. 3. ac Stark splitting under a resonant ($\Delta=0$) strong stochastic field in double optical resonance. The averaged population $\langle\rho_{33}\rangle$ of the third level is plotted as a function of the detuning Δ' of the probe field measured in units of Rabi frequency Ω. The dashed curves correspond to a PD field and the solid curves to a CH. The primed quantities Ω' and b' correspond to the probe field, and κ_3 is the spontaneous decay width of level $|3\rangle$. All widths are given in inverse seconds.

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Theory of Pure Rotational Transitions in Doubly Degenerate Torsional States of Ethane

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Pure rotational transitions in doubly degenerate torsional states of C_2H_6, with selection rules $\Delta K=0$, $\Delta J=0,\pm1$, are shown to be made allowed by Coriolis interaction between torsion and dipole-allowed vibrations. Expressions for integrated intensities are presented from which strengths of lines in the millimeter region are calculated.

Pure rotational transitions of highly symmetric molecules containing no permanent dipole moment has been, until recently, considered to be forbidden.[1] It has been shown theoretically that vibration-rotation interaction induces otherwise-forbidden rotational transitions in molecules, provided that they contain no center of symmetry.[2-4] Such forbidden transitions have been observed, with use of a variety of experimental techniques, in tetrahedral molecules[5] such as CH_4, and in symmetrical tops[6] such as AsH_3 and PH_3.

Ethane, C_2H_6, is generally thought of as containing a center of symmetry and belonging to the point group D_{3d}, corresponding to the equilibrium configuration of the rigid molecule. The hindered internal rotation, allowing tunneling from one equilibrium position to another, is not included as a symmetry operation in the group D_{3d}. Hougen[7] has shown that the proper symmetry group which takes into account all the feasible permutations and permutation inversions in ethane (which includes tunneling) is G_{36}[†]. According to this group, ethanelike molecules contain no center of symmetry and pure rotational transitions are therefore not strictly forbidden.

It is the purpose of this Letter to establish that pure rotational transitions in ethane are made active via an effective dipole moment arising from torsion-vibration-rotation interaction, and to present selection rules and expressions for the integrated intensities of such transitions. An estimate of the strength of lines in the microwave region will be given.

The interactions giving rise to these forbidden transitions are identical to those inducing the otherwise-forbidden pure torsional transitions observed in C_2H_6 (Ref. 8) and C_2F_6 (Ref. 9). These interactions, which have been discussed by Eggers[10] and by Eggers, Lord, and Wickstrom,[9] arise from the x,y-type Coriolis interaction coupling the torsion with the dipole-allowed doubly degenerate vibrations, and the z-type Coriolis interaction coupling the torsion with dipole-allowed nondegenerate vibrations. The selection rules and the corresponding transition intensities can be derived from an effective dipole moment in the space-fixed frame, given by

$$\mu_\omega = \sum_{\omega'} D_{\omega\omega'}^{(1)*} \tilde{\mu}_{\omega'}, \tag{1}$$

$$= P_\gamma \sum_{\omega'} D_{\omega\omega'}^{(1)*} \mu_{\omega'}{}^T J_{\omega'} \quad (\omega, \omega' = 0, \pm 1),$$

where $\tilde{\mu}_\omega$ is the effective dipole moment in the molecule-fixed frame, $D_{\omega\omega'}^{(1)*}$ are the rotational matrices, J_ω and $P\gamma$ are the components of the rotational and torsional angular momentum, respectively, γ is the torsional coordinate, and $\mu_\omega{}^T$ are coupling constants that obey the relation $\mu_1{}^T = \mu_{-1}{}^T \neq \mu_0{}^T$.

The form of the dipole moment in Eq. (1) can be derived from symmetry considerations alone. However, the dependence of the constants on the molecular parameters can be obtained by applying a contact transformation[11] that removes the off-diagonal Coriolis terms from the first-order Hamiltonian to the vibrational dipole moment. For example, a contact transformation that removes off-diagonal terms from the Hamiltonian of the type

$$-2A_\alpha \zeta_{4,k}{}^\alpha Q_{k\alpha} J_\alpha P_\gamma \quad (\alpha = x, y, z),$$

results in the constants $\mu_\omega{}^T$ given by

$$\mu_0{}^T = A \sum_{k=5,6} \zeta_{4,k}{}^z (1/\nu_k)(\partial \mu_z / \partial Q_k)_e,$$

