STARK EFFECT IN THE FIELD OF INCOHERENT RADIATION

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The Stark effect produced by intense resonant radiation is investigated. It is demonstrated that the radiation statistics are manifest in the structure and shape of the spectrum observed for the adjacent transition. It is found that from the shape of the spectrum one can distinguish between amplitude and frequency modulation of the radiation and derive from the experimental data fundamental information regarding the statistics of photons in an incoherent field.

INTRODUCTION

BONCH-Bruevich, et al.[1,2] investigated experimentally the Stark splitting of the absorption spectrum in the transition 1–2 under the influence of a high-power field in close resonance with the transition 2–3 (Fig. 1). The effect was interpreted under the assumption that the giant-pulse field can be regarded as monochromatic and its amplitude as constant. However, the smearing of the Stark structure in the case of resonant pumping was attributed in[1] to stochastic properties of the real laser pulse.

Thus, the effect as a whole, with allowance for the form of the spectrum and the width of its components, can be explained only by adequately describing the incoherent radiation causing the Stark effect. This, however, encounters considerable difficulties connected with the fact that the power spectrum alone, i.e., the second correlation moment of the random field, or even several higher moments, do not suffice to solve the problem. The Stark structure duplicates in explicit or in latent form the distribution of the amplitudes or frequencies of the radiation, and depends on their dispersion and correlation time, i.e., it incorporates the information contained in the entire infinite sequence of correlation moments. In other words, the statistics of the radiation becomes manifest in the Stark effect more completely and in greater variety than in interference phenomena investigated with the aid of several counters[3], and can be studied in a much better fashion.

The Stark effect has already been used for this purpose to investigate the statistics of electric fields in an equilibrium[4] or turbulent[5] plasma, and a similar application of this effect to the radiation field is highly promising. However, an all-out description of the Stark effect requires more complete information concerning the structure of the laser-radiation field than is at our disposal at present. It is therefore natural to use the opposite approach; we first investigate the phenomenon using models that admit of an exact solution, form a general ideal concerning the effect, and by comparing it with the observed one extract the required information from the experimental data.

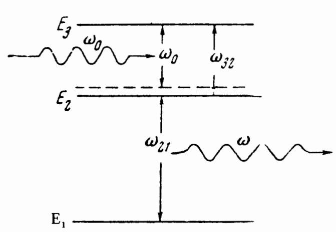

FIG. 1. Stark effect in a three-level system (dashed–virtual state not perturbed in the interaction with E_2).

A convenient model that admits of an all-out investigation of this kind is radiation whose phase, frequency, and amplitude constitute a randomly time-varying Markov process. The action of such a process on a two-level system was investigated in the past years[6] and, in particular, the changes of the populations ρ_{ii} and the phase elements ρ_{ik} determining the Stark effect on the same transition[6,7] were obtained under resonance conditions. An important advantage of investigating the Stark effect on an adjacent transition under conditions when the excited states are not populated is the fact that a description of the effect can be obtained by averaging the $\hat{S}$ matrix instead of the density matrix. This simplifies greatly both the solution of the problem and, in particular, its analysis. A realization of this methodological advantage has made it possible to cope, within the scope of a single article, with all the conceivable situations and to compare the spectra produced under phase, frequency, and amplitude-phase modulation of the radiation. It is remarkable that all the main results pertaining to the limiting cases have a quite general character, i.e., they do not depend on the chosen model of the random process, the specific features of which are revealed only in the transition regions. This means that the Stark effect can be utilized reliably to establish the type of modulation of the radiation and its statistical characteristics.

1. FORM OF SPECTRUM

The absorption of the probing monochromatic light in the transition 1–2 is determined by the correlation-theory formula

$$W_{21} = 2|B|^2 \operatorname{Re} \int_0^\infty \overline{\langle 1|\hat{d}(t)\hat{d}(0)|1\rangle} e^{i\omega t}\, dt, \tag{1.1}$$

where W_{21} is the transition probability, $\hat{d}$ the dipole moment of the transition, $\hbar = 1$, B is the amplitude of light frequency ω close to ω_{21}, and the superior bar denotes time averaging. Assuming that the time dependence of $\hat{d}(t)$ is due only to the action of the pump field on the 2–3 transition, which is not in resonance with the 1–2 transition, we can neglect the perturbation of level 1 ($S_{11} = 1$), and then

$$W_{21}=2|B|^2|d_{12}|^2\operatorname{Re}\int_0^\infty\overline{S_{22}(t)}e^{i\omega t}dt=2\pi|d_{12}|^2|B|^2G(\omega),\quad(1.2)$$

where $\hat{S}(t)$ obeys the Schrödinger equation

$$d\hat{S}/dt=-i\hat{H}\hat{S}=-i[\hat{H}_0-\hat{d}E(t)]\hat{S}.\quad(1.3)$$

with a Hamiltonian that depends randomly on the time. This dependence is due to the stochastic properties of the electromagnetic field of the illumination

$$E(t)=E_0(t)\exp\left\{-i\omega_0t-i\int_0^t\Delta\omega(t')dt'-i\theta(t)\right\},\quad(1.4)$$

The amplitude, frequency shift, and phase of which are assumed henceforth to be a purely discontinuous Markov stationary process.

The last assumption makes it possible to write down immediately an equation[8] for the time behavior of the $\hat{S}$-matrix density:

$$i\frac{\partial\hat{S}(\alpha,t)}{dt}=\hat{H}(\alpha,t)\hat{S}-\frac{i}{\tau_0}\left[\hat{S}-\int f(\alpha,\alpha')\hat{S}(\alpha',t)d\alpha'\right],\quad(1.5)$$

in which α is the Markov variable (or a set of simultaneously changing variables), which imparts a random character to the time dependence of the Hamiltonian $H(\alpha(t), t)$. The initial condition for (1.5) is specified by the natural property of the $\hat{S}$ matrix, namely that it becomes equal to unity at $t = 0$, as a result of which its density at that instant coincides with the distribution of the random quantity

$$\hat{S}(\alpha,0)=\varphi(\alpha),\qquad\int\varphi(\alpha)d\alpha=1.\quad(1.6)$$

The kernel of the integral equation (1.5), $f(\alpha, \alpha')$, is the density of the conditional distribution that specifies the probability of variation of α' by α at instants that have a Poisson distribution with a mean spacing between them τ_0. The completely averaged $\hat{S}$ matrix which enters in (1.2) is

$$\overline{S}=\int\hat{S}(\alpha,t)d\alpha,\quad(1.7)$$

so that the normalized contour of the spectrum can be expressed directly in terms of the Fourier transform of the solution of (1.5):

$$G(\omega)=\int d\alpha\frac{1}{\pi}\operatorname{Re}\int_0^\infty S_{22}(\alpha,t)e^{i\omega t}dt=\frac{1}{\pi}\operatorname{Re}\int d\alpha Q_{22}(-i\omega,\alpha).\quad(1.8)$$

2. PHASE MODULATION

In the simplest case when E_0 = const and $\Delta\omega = 0$, the only source of pump-radiation spectrum broadening is the random drift of its phase $\theta(t)$. Eliminating the explicit dependence on the time in (1.5) by means of the transformation $S_{32} = S_{32}^{v}\exp(-i\omega_0 t)$, we have

$$i\frac{dS_{22}}{dt}=\omega_{21}'S_{22}+e^{i\theta}V_{23}S_{32}^{v}-\frac{i}{\tau_0}\left[S_{22}-\int f(\theta-\theta')S_{22}(\theta')d\theta'\right],\quad(2.1a)$$

$$i\frac{dS_{32}^{v}}{dt}=(\omega_{31}'-\omega_0)S_{32}^{v}+e^{-i\theta}V_{32}S_{22}-\frac{i}{\tau_0}\left[S_{32}^{v}-\int f(\theta-\theta')S_{32}^{v}(\theta')d\theta'\right],\quad(2.1b)$$

where $\omega_{21}' = \omega_{21} - i\Gamma_{21}$, $\omega_{31}' = \omega_{31} - i\Gamma_{31}$, Γ_{21} and Γ_{31} are the widths of the corresponding transitions, $V_{23} = E_0 d_{23}$, and $f(\theta - \theta')$ is assumed to be a function of the phase difference only.

Multiplying the second equation by $e^{i\theta}$ and integrating term by term with respect to θ, we obtain

$$id\overline{S}_{22}/dt=\omega_{21}'\overline{S}_{22}+V_{23}S_{32}^{v}(1,t),\quad(2.2a)$$

$$idS_{32}^{v}(1,t)/dt=(\omega_{31}'-\omega_0)S_{32}^{v}(1,t)+V_{32}\overline{S}_{22}-i\Gamma S_{32}^{v}(1,t),\quad(2.2b)$$

where

$$S_{32}^{v}(1,t)=\int e^{i\theta}S_{32}^{v}(t,\theta)d\theta,\quad(2.3a)$$

$$\Gamma=\frac{1}{\tau_0}\operatorname{Re}\left[1-\int e^{i\theta}f(\theta)d\theta\right];\quad(2.3b)$$

Γ is the width of the acting radiation, which has a Lorentz form and whose frequency shift is included in ω_0[8].

Taking the Laplace transform of (2.2) and solving these equations with respect to $\overline{Q}_{22}$, we obtain ultimately

$$G(\omega)=\frac{1}{\pi}\operatorname{Re}i\left[(\omega-\omega_{21}')-\frac{V^2}{\omega-\omega_{31}'+\omega_0+i\Gamma}\right]^{-1}$$
$$=\frac{1}{\pi}\frac{\Gamma_{21}+W(\omega)}{[\omega-\omega_{21}-W(\omega-\omega_{31}+\omega_0)/(\Gamma+\Gamma_{31})]^2+[W(\omega)+\Gamma_{21}]^2}\quad(2.4)$$

$$W(\omega)=\frac{V^2(\Gamma+\Gamma_{31})}{(\omega-\omega_{31}+\omega_0)^2+(\Gamma+\Gamma_{31})^2},\qquad V=\gamma\overline{|V_{23}|^2}.\quad(2.5)$$

Formulas (2.4) show that a quadratic Stark effect is realized at a relatively weak illumination power $W \ll \Gamma + \Gamma_{31}$. The spectrum consists of two Lorentz lines, single-photon and two-photon, with resonances at the respective frequencies $\omega_1 = \omega_{21} + \delta$ and $\omega_2 = \omega_{31} - \omega_0 - \delta$, where

$$\delta=\frac{V^2(\omega_0-\omega_{32})}{(\omega_0-\omega_{32})^2+(\Gamma+\Gamma_{31})^2},\quad(2.6)$$

and the widths and integral intensities of these lines are given by the formulas

$$\gamma_1=W(\omega_{21})+\Gamma_{21},\quad I_1=1-\delta/(\omega_0-\omega_{32}),$$
$$\gamma_2=\Gamma+\Gamma_{31}+W(\omega_{21})\approx\Gamma+\Gamma_{31}\quad I_2=\delta/(\omega_0-\omega_{32}).\quad(2.7)$$

In the case of powerful illumination, $W \gg \Gamma + \Gamma_{31}$, the Stark effect becomes linearized and the spectrum consists of two Lorentz lines of equal intensity, disposed symmetrically about ω_{21} at a distance V, and having a width $(\Gamma + \Gamma_{31} + \Gamma_{21})/2$.

As seen from (2.7), observation of a single-photon line is easiest not only because its integral intensity is larger, but also because it remains narrower than the two-photon line so long as the proper widths of the transitions are negligibly small ($\Gamma_{21}, \Gamma_{31} \ll \Gamma$). However, strong broadening of the second level can greatly influence the situation and even reverse it. The point is that the broadening affects only the single-photon line and not the two-photon line, and when $\Gamma_{21} \gg \Gamma$ it can even offset the intensity advantage, as was noted in[1].

3. FREQUENCY MODULATION

Let now E_0 = const and θ = const, and let $\delta\omega$ be a randomly varying frequency deviation distributed in any time interval in accordance with the law $\varphi(\delta\omega)$

about ω_0. The spectrum of such radiation is given by the formulas

$$g(\omega) = -\pi^{-1} \operatorname{Im} \Phi(\omega),$$
$$\Phi(\omega) = -i \int_0^\infty \exp \overline{\left\{ i\omega_0 t + i \int \delta\omega \, dt' - i\omega t \right\}} \, dt \tag{3.1}$$

and its general form is well known[8]. If, in particular, the frequency is detuned in an uncorrelated manner, then

$$\Phi(\omega) = \frac{j(\omega)}{1 - i\tau_0^{-1} j(\omega)}, \quad j(\omega) = \int \frac{\varphi(z)\, dz}{\omega_0 + z - \omega + i\tau_0^{-1}}. \tag{3.1a}$$

so long as the frequency dispersion $\Delta = (\overline{\delta\omega^2})^{1/2}$ is much larger than the rate of its change τ_0^{-1}, the shape of the spectrum duplicates the distribution $\varphi(\delta\omega)$ (quasistatic contour), but when $\Delta\tau_0 \ll 1$, then the spectrum narrows down to a Lorentz contour of width $\Delta^2\tau_0$ (narrowed contour).

The Stark effect produced by such radiation is sensitive not only to the scale of the parameter $\Delta\tau_0$, but also to the interaction strength $V\tau_0 = d_{23}E_0\tau_0$, and to the relative values of these parameters. Assuming that

$$f(\delta\omega, \delta\omega') = \varphi(\delta\omega), \tag{3.2}$$

which was already implied in (3.1a), we can solve the problem in the most general form and facilitate by the same token the analysis of all the situations that are possible here. This assumption, at the same time, is not a considerable restriction on the problem, since in practice all the results can be reduced to a form that is independent of the model of the modulation, and their generality can be verified.

A physically important limitation in this case, as in the preceding one, is the assumption that the radiation has a single-mode character; this assumption follows from the assumptions made here and from the temporal field variation postulated in (1.4). Nonetheless, such a structure of the laser radiation is one of the most important and frequently discussed models[3], and we shall therefore deal only with this structure, and relegate the analysis of the multimode model to the next section.

In the representation where

$$S_{32} = S_{32}^v \exp\left[-i\omega_0 t - i \int \delta\omega \, dt \right],$$

Eqs. (1.5), with allowance for (3.2), can be reduced to the form

$$i dS_{22} / dt = \omega_{21}' S_{22} + V_{23} S_{32}^v - i\tau_0^{-1} [S_{22} - \varphi(\delta\omega) \bar{S}_{22}], \tag{3.3a}$$

$$i dS_{32}^v / dt = (\omega_{31}' - \omega_0 - \delta\omega) S_{32}^v + V_{32} S_{22} - i\tau_0^{-1} [S_{32}^v - \varphi(\delta\omega) \bar{S}_{32}^v], \tag{3.3b}$$

where $V_{23} = E_0 d_{23} e^{i\theta}$. Taking the Laplace transforms of these equations, we obtain

$$ipQ_{22} - i\varphi(\delta\omega) = (\omega_{21}' - i\tau_0^{-1}) Q_{22} + i\tau_0^{-1} \bar{Q}_{22} \varphi(\delta\omega) + V_{23} Q_{32}^v, \tag{3.4a}$$

$$ipQ_{32}^v = (\omega_{31}' - \omega_0 - \delta\omega - i\tau_0^{-1}) Q_{32}^v + V_{32} Q_{22} + i\tau_0^{-1} + \bar{Q}_{32}^v \varphi(\delta\omega). \tag{3.4b}$$

Solving this system with respect to Q_{22} and Q_{32}^V and integrating the results with respect to $\delta\omega$ and again solving with respect to the average values $\overline{Q}_{22}$ and $\overline{Q_{32}^V}$, we obtain from (1.8) after a number of transformations

$$G(\omega) = \frac{1}{\pi} \operatorname{Re} \frac{j_1 - i\tau_0^{-1} j_2}{1 - \tau_0^{-1} j_1 - \tau_0^{-1} (\omega - \omega_{21}') j_2}, \tag{3.5}$$

where

$$j_1 = i \int \frac{\varphi(z)\, dz}{\omega - \omega_{21}' + i\tau_0^{-1} - W(\omega, z)},$$
$$j_2 = i \int \frac{\varphi(z)\, dz}{(\omega - \omega_{31}' + \omega_0 + z + i\tau_0^{-1})(\omega - \omega_{21}' + i\tau_0^{-1} - W)} \tag{3.6}$$

and

$$W(\omega, z) = \frac{V^2}{\omega - \omega_{31}' + \omega_0 + z + i\tau_0^{-1}}. \tag{3.7}$$

Since both integrals j_1 and j_2 are expressed in terms of $V^2 = |V_{23}|^2$, the form of the Stark spectrum does not depend on the phase distribution over the different frequencies ($\theta(\delta\omega)$), regardless of its character.

Formulas (3.5), (3.6) and (3.7) give the general solution of the problem in the entire region of its definition, shown in Fig. 2. This region, naturally, breaks up into six different segments, thereby facilitating somewhat the analysis of the general expressions, an analysis which in general is far from simple. To gain a complete idea of the concrete structure of the spectrum and its transformation, we consider in sequence all six different limiting situations and the most important transitions between them.

We start with a situation wherein the frequency of the acting light is modulated very slowly: $\tau_0^{-1} \ll V, \Delta$ (the quadrant A and B in Fig. 2). In this region, the general formula (3.5) greatly simplifies and assumes the form

$$G(\omega) = \pi^{-1} \lim_{\tau_0 \to \infty} \operatorname{Re} j_1. \tag{3.8}$$

Carrying out integration in (3.6) and taking the limit in accordance with this recipe, we can easily establish that the spectrum has a Stark structure and that its form (neglecting $\Gamma_{21}, \Gamma_{31} \ll V, \Delta$ at resonant pumping) is given by the following function:

$$G(\Omega) = \frac{V^2}{\Omega^2} \varphi\left(\frac{V^2}{\Omega} - \Omega\right), \tag{3.9}$$

where $\Omega = \omega - \omega_{21}$ is the deviation, in terms of frequency, from the position of the unsplit line.

In region A, when the pump field power is so large

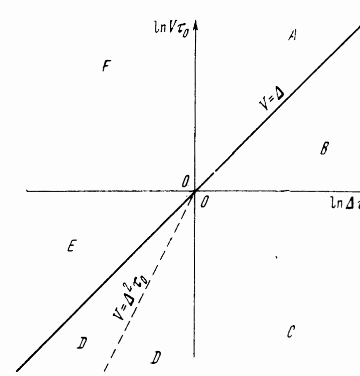

FIG. 2. Region where the solution is defined for resonant pumping with frequency-modulated radiation (the dashed line indicates the boundary between the linear and quadratic effects in the region DD).

that $V \gg \Delta$, the effect should obviously be linear. Indeed, according to (3.9), both Stark components, separated in the spectrum by an amount 2V, have shapes that duplicate the frequency distribution of the acting light, but with half the width (A in Fig. 3):

$$G_{1,2}(\Omega) = {}^1/_2\varphi[2(\Omega \pm V)]. \tag{3.9a}$$

On the other hand, if the pump field is weaker, $V \ll \Delta$ (region B on Fig. 2), then a quadratic Stark effect appears in the spectrum: in the center of the line with total width $\sim V^2/\Delta$ there is observed an even narrower gap with width of the same order (B in Fig. 3). Such an unusual shape can easily be interpreted by recognizing that the center of this spectrum is made up of single-photon components shifted in both directions by an amount equal to the quadratic shift, and that its periphery is made up of two-photon components that decrease rapidly in intensity.

Let now $V \ll \Delta$, τ_0^{-1}. To facilitate the analysis of this situation, it is useful to reduce (3.5) by identity transformations to the following more convenient form:

$$G(\omega) = \frac{1}{\pi}\operatorname{Re}\frac{i}{\omega - \omega_{21}' - V^2F(\omega)}, \tag{3.10}$$

where

$$F(\omega) = \frac{I(\omega)}{1 - [i\tau_0^{-1} - \gamma(\omega)]I(\omega)}; \tag{3.11}$$

$$I(\omega) = \int\frac{\varphi(z)\,dz}{\omega - \omega_{31}' + \omega_0 + z + i\tau_0^{-1} - \gamma(\omega)}, \quad \gamma(\omega) = \frac{V^2}{\omega - \omega_{21}' + i\tau_0^{-1}}. \tag{3.12}$$

In addition, it is appropriate to confine the analysis to the condition $\Gamma_{31}, \Gamma_{21} \ll \tau_0^{-1}$, since when this condition is inverted everything reduces to the result given in (3.8). Further, since the smallest quantity in the situation under consideration is the interaction with the light, it is easily seen that the frequency dependence of $F(\omega)$ is weakly pronounced within the limits of the resonance, since $\gamma(\omega)$ is small. On this basis we can replace $F(\omega)$ by its resonant value and thus extract immediately from (3.10) the form of the single-photon line:

$$G_1(\omega) = \frac{1}{\pi}\operatorname{Re}\frac{i}{\omega - \omega_{21}' - V^2F(\omega_{21})}. \tag{3.13}$$

It turns out that, regardless of the form of the acting spectrum ($\Delta\tau_0 \gtrless 1$), it constitutes a Lorentz contour with the respective width and shift,

$$W = -V^2\operatorname{Im}\Phi(\omega_{32} - i\Gamma_{31}), \quad \delta = V^2\operatorname{Re}\Phi(\omega_{32} - i\Gamma_{31}), \tag{3.13a}$$

the determination of which generalizes directly the formulas (2.5) and (2.6). At $\Gamma_{31} = 0$ it is easy to recognize in these formulas the usual result of perturbation theory, and this is perfectly legitimate: so long as the radiation spectrum width is larger than the probability of the relaxation induced by the radiation, the relaxation has a kinetic (exponential) character, which can lead only to a broadening and a shift of the line. This is precisely what takes place within the range of validity of (3.13).

In region C (Fig. 2), in particular where $g(\omega) = \varphi(\omega - \omega_0)$, we obtain a result that does not depend on τ_0:

$$W = \pi V^2\varphi(\omega_{32} - \omega_0), \quad \delta = \pi V^2 \mathrm{P}\int\frac{\varphi(z)\,dz}{\omega_0 - \omega_{32} + z}, \tag{3.14}$$

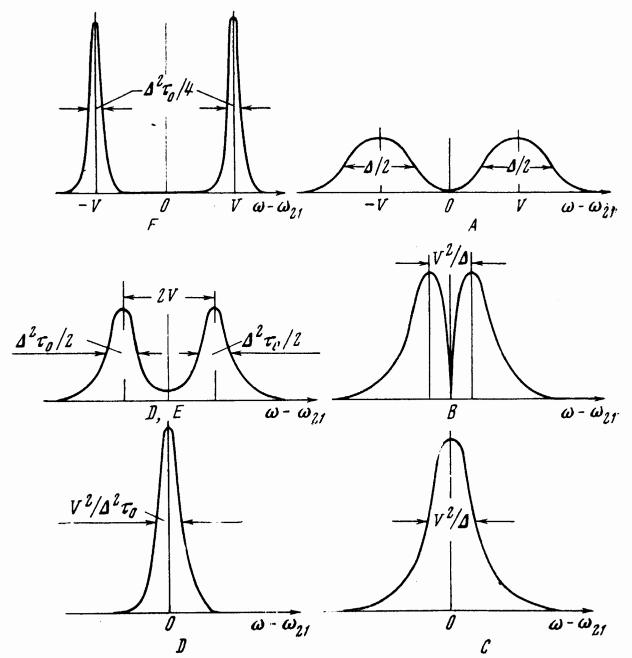

FIG. 3. Spectra of Stark effect in the corresponding segments of Fig. 2.

and this is natural, since the radiation spectrum is a quasistatic contour[1]. Nonetheless, there is a strong qualitative difference between the quadratic Stark effect of this type (C in Fig. 3) and that observed in the preceding case (B in Fig. 3); this difference is expressed in the presence or absence of a dip in the center of the spectrum. In the level energy structure produced by the powerful light, this dip is the phase analog of the Bennett dip in the population spectrum, which arises under analogous conditions. Its appearance at $V\tau_0 = 1$ can serve as a good method of determining τ_0 under conditions when this quantity does not enter directly either into the spectrum of the acting light or into the form of the Stark effect induced by it.

We now proceed to consider the Stark effect produced under the action of a frequency-modulated wave with Lorentz spectrum, i.e., all the remaining cases, unified by the condition $\Delta\tau_0 \ll 1$. It turns out that this condition suffices to obtain a single lucid expression for them. To this end, we represent (3.11) in the form

$$F(\omega) = [I^{-1}(\omega) - i\tau^{-1}(\omega) + \operatorname{Re}\gamma(\omega)]^{-1}, \tag{3.15}$$

where

$$I(\omega) = \int\frac{\varphi(z)\,dz}{[\omega - \omega_{31}' + \omega_0 - \operatorname{Re}\gamma(\omega)] + z + i\tau^{-1}(\omega)}, \tag{3.16}$$

and

$$\frac{1}{\tau(\omega)} = \frac{1}{\tau_0} - \operatorname{Im}\gamma(\omega) = \frac{1}{\tau_0} + \frac{V^2\tau_0}{\tau_0^2(\omega - \omega_{21})^2 + 1}. \tag{3.16a}$$

Using the condition $\Delta \ll \tau_0^{-1} \le \tau^{-1}(\omega)$, and also the possibility of confining ourselves to a near-resonance region $|\omega - \omega_{31} + \omega - \operatorname{Re}\gamma(\omega)|\,\tau_0 \ll 1$ of sufficient width, we can expand $I(\omega)$ in a series in both small parameters and obtain as a result a much simpler expression, which is valid when $V\tau_0 \gg 1$ and $V\tau_0 \ll 1$:

[1]The condition for the applicability of (3.13) is certainly satisfied here, since $W < \pi V^2\varphi(0) \approx V^2/\Delta \ll \Delta$.

$$F(\omega) = [\omega - \omega_{31}' + \omega_0 + i\Delta^2\tau(\omega)]^{-1}. \quad (3.17)$$

Let now $V\tau_0 \ll 1$ (regions D and E in Fig. 2), and suppose that both resonances fall in the frequency region $|\omega - \omega_{21}| \ll \tau_0^{-1}$. Then

$$\tau(\omega) = \tau_0, \quad (3.18)$$

and the dependence on the frequency in (3.17) becomes trivial. Moreover, substitution of this result in (3.10) reduces this equation to the same form as (2.4), with

$$\Gamma = \Delta^2\tau_0. \quad (3.19)$$

This coincidence is not accidental. The phase modulation can be regarded as frequency modulation by a process of a different type (by collision). Consequently, wherever the perturbation of the frequency is small, i.e., within the limits of perturbation theory with respect to the parameter $\Delta\tau_0$, the results should coincide not only in form but also in essence including satisfaction of Eq. (3.19). Therefore if we regard the width of the spectrum of the acting light as a phenomenological parameter, then formulas (2.4) and (2.5) acquire a perfectly general character, which is independent not only of the type of frequency modulation but of whether it is weak or strong: they remain valid also in the case of a full phase change ($\Gamma = \tau_0^{-1}$), i.e., beyond the limits of the coincidence of (2.3) and (3.19). The limitations on the generality concern only the form of the spectrum, which should have a shock profile ($\Delta\tau_c \ll 1$), and the radiation power ($V\tau_c \ll 1$; here τ_c is the field-frequency correlation time). Therefore the spectra pertaining to this region (D and E in Fig. 3) duplicate the Stark structure at resonant pumping by radiation, regardless of whether it is the frequency that is detuned or the phase that changes. Still within the region D, at $V\tau_0 = (\Delta\tau_0)^2$, the Stark effect becomes linear, and with further increase of the light intensity the width of its components becomes independent of the intensity. The limit of the quadratic effect is shown dashed in Fig. 2.

Let, finally, $V \gg \Delta^2$, but $\Delta\tau_0 \ll 1$ as before, corresponding to regions E and F in Fig. 2. In both cases we deal with a linear Stark effect, i.e., the resonances are located at the frequencies $\omega = \omega_{21} \pm V$, and their width, as shown by special investigation, is

$$\gamma = \frac{\Delta^2\tau_0}{2}, \quad \frac{1}{\tau} = \frac{1}{\tau_0} + \frac{2V^2\tau_0}{1 + 2V^2\tau_0^2}. \quad (3.20)$$

When $V\tau_0 \ll 1$ we obtain from this the already described result for the region E, namely formula (3.18) and the corresponding width (3.19). On the other hand, when $V\tau_0 \gg 1$, then $\tau = \tau_0/2$ and

$$G_{1,2} = \frac{1}{\pi}\operatorname{Re}\frac{i}{\omega - \omega_{21} \pm V + i\Delta^2\tau_0/4}, \quad (3.21)$$

i.e., the spectrum retains the previous form, but becomes half as wide. In the intermediate region the width changes smoothly as a function of the power, in accordance with (3.20). Thus, here too, as in the transition from B to A, the very onset of narrowing can serve as a criterion for the measurement of the quantity $\tau_0 \sim V^{-1}$, thus affording an apparently unique possibility, in principle, of separating the two independent parameters Δ and τ_0, which enter multiplicatively in the experimentally observed spectrum width $\Gamma = \Delta_2\tau_0$.

In conclusion, it is useful to call attention to the similarity between the results obtained at the largest $V \gg \Delta, \tau_0^{-1}$, i.e., in the regions F and A. It is easy to establish a single formula describing in continuous fashion the transition between these limiting cases, in spite of the fact that the contour of the Stark components is incidentally transformed from the Lorentz to the static type. Recognizing that at very large V the energy of the second level, calculated in first order in $\delta\omega$, is $\epsilon_2^{1,2} = \omega_{21} - \frac{1}{2}\delta\omega \pm V$, we can obtain directly from (1.2)

$$G_{1,2}(\omega) = \frac{1}{\pi}\operatorname{Re}\int_0^\infty \exp\left\{-i\int_0^t \varepsilon_2^{1,2}dt'\right\}e^{i\omega t}\,dt. \quad (3.22)$$

Comparing this result with the definition given for $g(\omega)$ in (3.1), we get

$$G_{1,2}(\omega) = g(\omega - \omega_{21} \pm V, \Delta/2), \quad (3.23)$$

which indeed agrees with the limiting formulas (3.9a) and (3.21), which are obtained from this at $\Delta\tau_0 \gg 1$ and $\Delta\tau_0 \ll 1$, respectively, under resonant pumping conditions. This shows that the results pertaining to both cases have a general character and do not depend on the assumed model of frequency modulation.

Summarizing the foregoing, we can conclude that everywhere except in the regions D and E of Fig. 2 the Stark spectra induced by frequency-modulated radiation differ from those occurring in a phase-modulated field. But even when the spectra in the regions D and E are outwardly similar, by simply increasing the power and changing over into the region F it is possible to distinguish one type of modulation from the other. It is also remarkable that regardless of whether the spectrum of the acting pump field is quasistatic (right half-plane, Fig. 2) or narrowed (left half-plane), it is possible, by varying its power, to obtain three Stark spectra of different structures and to extract from them complete information on the parameters Δ and τ_0, including also the form of the frequency distribution $\varphi(\delta\omega)$ when the latter is significant.

4. AMPLITUDE-PHASE MODULATION

By the same method as in the preceding section, it is easy to consider the case of pure amplitude modulation ($\delta\omega = 0$, $\theta = \text{const}$, $E = E_0(t)$). It is difficult to visualize, however, a mechanism capable of producing this type of modulation, since nonadiabatic processes that change the radiation amplitude should affect to an equal degree also its phase. We shall therefore focus our attention on modulation such that both the amplitude and the phase of the radiation are changed simultaneously and without correlation, i.e., $\delta\omega = 0$, and

$$f(E_0, \theta; E_0', \theta') = \varphi(E_0)/2\pi. \quad (4.1)$$

As a result we obtain from (1.5), by making the substitution $S_3 = S_{32}^V \exp(-i\omega_0 t)$

$$i\frac{dS_{22}}{dt} = \omega_{21}S_{22} + d_{23}E_0e^{i\theta}S_{32}^v - \frac{i}{\tau_0}\left[S_{22} - \frac{\varphi(E_0)}{2\pi}\bar{S}_{22}\right], \quad (4.2a)$$

$$i\frac{dS_{32}^v}{dt} = (\omega_{31} - \omega_0)S_{32}^v + d_{32}E_0e^{-i\theta}S_{22} - \frac{i}{\tau_0}\left[S_{32}^v - \frac{\varphi(E_0)}{2\pi}\overline{S_{32}^v}\right]. \quad (4.2b)$$

Solving this system by the method used for (3.3), we get

$$G(\omega)=\frac{1}{\pi}\operatorname{Re}\frac{j_1}{1-i\tau_0^{-1}j_1}, \quad (4.3)$$

where

$$j_1=i\int\frac{\varphi(E_0)\,dE_0}{(\omega-\omega_{21}+i\tau_0^{-1})-W(\omega,E_0)}, \quad (4.4)$$

$$W(\omega,E_0)=\frac{|d_{23}|^2E_0^2}{\omega-\omega_{31}+\omega_0+i\tau_0^{-1}}. \quad (4.5)$$

In the weak-interaction region, $V^2 = |d_{23}|^2\overline{E_0^2} \ll \tau_0^{-2}$, a quadratic Stark effect is realized, similar to that produced under the influence of radiation modulated in a different manner. For resonant pumping, for example, the spectrum does not differ in any way from that shown in Fig. 3C, with the exception of the width, which equals $V^2\tau_0 = |d_{23}|^2\overline{E_0^2}\tau_0$. A sharp qualitative difference is observed only in the case of strong interaction, when the effect becomes linearized.

$$G(\omega)=\frac{1}{\pi}\lim_{\tau_0\to\infty}\operatorname{Re}j_1=\frac{1}{2|d_{23}|}\left[\varphi\left(\frac{\omega-\omega_{21}}{|d_{23}|}\right)+\varphi\left(\frac{\omega_{21}-\omega}{|d_{23}|}\right)\right] \quad (4.6)$$

and the form of both components duplicates not the spectrum of the acting radiation (of Lorentz form with width τ_0^{-1}), but the amplitude distribution $\varphi(E_0)$. Therefore the best method of distinguishing amplitude-modulated radiation from any other radiation is to compare the width and form of the Stark components with the corresponding characteristics of the pump light. In the case of sufficiently strong illumination, the Stark components should be broader and different in form, as was indeed observed[1]. In addition, with increasing light intensity, the transition from the quadratic effect to the linear is effected directly, bypassing the intermediate stage produced before or after the transition (B and E of Fig. 3) in the case of frequency modulation.

It is important to emphasize that the result (4.6) is perfectly general: it does not depend on the type of the random process modulating the amplitude, and occurs whenever $V^2\tau_c^2 \gg 1$, where τ_c is the field-amplitude correlation time. Therefore the linear Stark effect is the most direct method of reconstructing the distribution of the amplitudes in the field of the acting radiation. If, in particular, we deal with simultaneous emission of a multimode laser, it is natural to expect, in view of the fundamental limit theorem, that the amplitude of the summary field has a Rayleigh distribution

$$\varphi(E_0)=2\frac{E_0}{D^2}\exp\left(-\frac{E_0^2}{D^2}\right), \quad D=\overline{E_0^2}. \quad (4.7)$$

In such a case the splitting of the line into two components can readily be observed experimentally (Fig. 4a).

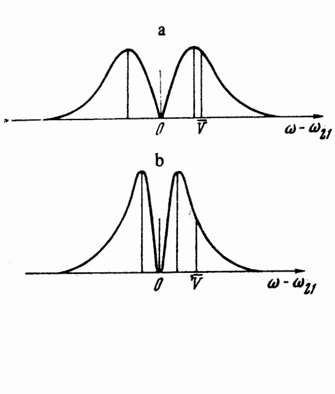

FIG. 4. Possible form of resonant Stark effects when all the modes are radiated simultaneously (a) and at different times (b).

Indeed, in the case of resonant action of a giant pulse, a split line was observed, whose width, however, greatly exceeded the width of the acting radiation[1], so that it could not be anything but the linear effect in an amplitude-modulated field. An analysis of all the results given above shows that the smearing of such a broad dip in the spectrum (4.6) cannot be due either to the simultaneous action of other types of modulation or to the proper broadening of the atomic line, since all these widths, in accordance with the measurements, were smaller by one order of magnitude than the dip width V due to (4.7). Judging from this, we are dealing here not with a Rayleigh distribution and consequently not with simultaneous radiation of all modes, but more readily with pulsed radiation of the modes at different times. The light pulses act individually on the atom, one after the other, and in the intervals between them the field weakens to the extent that a quadratic Stark effect is observed, i.e., a practically unshifted line filling the breach in the central part of the spectrum. In other words, the statistics of the summary field of such a structure is closer to the Holtsmark distribution[4], which has a narrow dip at the center (Fig. 4b) that can readily be washed out by any type of broadening.

CONCLUSION

As seen from the foregoing, the use of new sources of light of high intensity not only greatly broadens the range of research, but uncovers fundamentally new possibilities for coherence theory. Weak illumination leads only to a trivial Stark effect, causing the appearance of a two-photon line and a light-induced broadening of the single-photon line, which can be easily described within the framework of ordinary perturbation theory:

$$G(\omega)=\frac{1}{\pi}\operatorname{Re}\frac{i}{\omega-\omega_{21}'-R(\omega_{21})}$$

$$R(\omega)=|d_{23}|^2\int_0 \overline{E^*(t)E(0)}\exp\{-i(\omega-\omega_{31}')t\}\,dt=\delta-i\Gamma.$$

Knowledge of the second correlation moment, which can readily be reconstructed from the form of the emission spectrum, suffices completely to calculate by means of these formulas the shift quadratic in the field and the broadening of the Stark components. New and more detailed information on the statistics of the radiation field can be obtained from the Stark effect only by going beyond the limits of perturbation theory, i.e., in fields for which the inequality $V^2\tau_c^2 \gg 1$ is valid.

It was shown above that the spectra in this region (Fig. 4 and A, B, and F in Fig. 3) differed greatly from one another and from the field observed in the case of weak illumination. This makes it possible in principle a) to distinguish between frequency and amplitude modulation of the radiation, b) to measure the dispersion and the correlation time of the frequency in the case when they are inaccessible to direct observation, c) to determine the distribution of the radiation amplitude and to extract from it an idea of the possible structure of the light flux.

To facilitate the completion of this task, all the results pertaining to the pure limiting situations are presented in as general a form as possible. In this form they remain valid also in the presence of correlation between the discontinuities of the frequency and of the amplitude, and also when a change takes place in the type of the random process, provided τ_0 is replaced by the correlation time of the corresponding variable.

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