Two-quantum resonance excitation of two-level systems by stochastic fields

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The two-quantum excitation of a two-level system by broad-band radiation is investigated theoretically and experimentally. It is shown that the character and efficiency of the radiation's action can differ very greatly for fields that have different statistics. It is found that correlation between the harmonics of the spectrum can give an excitation efficiency not inferior to that of a monochromatic field at any level of excitation. Intraspectral correlation was produced experimentally by heterodyning, and it was shown that an originally inefficient signal having the statistics of complex Gaussian noise can be converted into an efficient correlated signal without any essential change of the spectrum, power, and intensity of the fluctuations as compared with the original signal. The action of the correlated field has special features combining those of monochromatic and broad-band nonmonochromatic excitation. In particular, the excitation spectrum of the correlated field is sharply resonant. The resonance part of the spectrum shows a paradoxical behavior: It becomes narrower and narrower as the width of the spectrum of the exciting field is increased.

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The study of the behavior of resonance systems in stochastic fields is of interest in various branches of physics. Such research is particularly significant for quantum radiation physics and nonlinear optics, because often the nonmonochromaticity of the field acting on resonant atomic-molecular and spin systems cannot be eliminated under actual conditions.

Experience in research on one-quantum and stepwise processes in classical resonant systems characterized by a single natural frequency has brought about a certain inertia in our ideas, with the supposition that under conditions close to resonance systems will be the more strongly excited, the narrower the spectrum of the applied radiation for a given intensity. It is intuitively assumed that the behavior of systems in stochastic fields becomes more and more like that found with monochromatic excitation, the narrower the spectrum of the nonmonochromatic field is made.

It is not hard to give examples of phenomena that do not fit into the framework of these ideas. It is well known[1] that under certain conditions nonmonochromatic fields can excite a resonance system $k!$ times more intensely than monochromatic fields of the same power (here k is the multiplicity of the many-photon resonance). Another example[2]: The broadening of the transition line in the quadratic dynamic Stark effect is greater the narrower the spectrum of the nonmonochromatic excitation. Both of these phenomena are caused by the fluctuations of the excitation's intensity and occur when the perturbation of the system is weak and the saturation effects are negligible. Moreover, these phenomena are brought about by the action of radiation with a narrow spectral band with an upper limit on its width imposed by definite conditions.[1,2]

Although these examples do correct the usual ideas about resonance phenomena in nonmonochromatic fields, it is still hard to expect monochromatic and broad-band stochastic radiations to be closely similar in their efficiencies and the nature of their effects on a system, at comparable intensities and without limitations in principle on the width of the excitation spectrum. In fact, if we consider, for example, a two-quantum process of resonance excitation of a two-level system by broad-band radiation (see Fig. 1), it can be seen that the main effect is given by the spectral region $\omega_0 + \omega_1 \approx \omega_{21} \pm \gamma_2$, where γ_2 is the line width of the transition, and ω_0 and ω_1 are the frequencies of the harmonics of the excitation, which run through all values in the spectrum. But since the main part of the radiation's intensity is concentrated in a band of width $\sim\Delta$, it is evident that for a given intensity the larger Δ, the smaller the excitation efficiency will be. Therefore it would seem that in any many-quantum resonance process broad-band radiation could not achieve the efficiency of a monochromatic field. This sort of argument leads to the conclusion that also the spectrum of the excitation in the process in question cannot be noticeably narrower than the pumping spectrum when $\Delta \gg 2$.

Nevertheless these conclusions are not absolutely correct. In this two-quantum process the action of broad-band radiation can be made no less efficient than monochromatic radiation. Furthermore there will then be a sharp resonance with a width considerably smaller than Δ. It will be shown later that the broad-band effect becomes both efficient and selective when its statistics satisfy definite requirements.

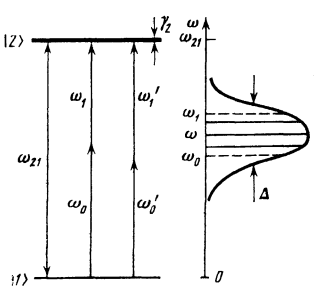

FIG. 1. Scheme of two-quantum excitation of a resonant system by nonmonochromatic radiation. To the right is shown the position of pairs of mutually complementary harmonics in the spectrum of the exciting radiation.

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The effect of the statistics of the radiation (both classical and quantum) on the two-quantum excitation process in a two-level system has been discussed repeatedly. The actions of classical fields having the statistics of complex Gaussian and of Gauss-Markov processes are nearly equally efficient.[3] Quantum fluctuations can cause a marked difference between the effects of thermal and laser radiations.[4,5] The essential influence of the quantum statistics of the radiation on the two-quantum excitation process was made apparent in Ref. 6. For weak perturbation of the system it was shown that the radiation resulting from spontaneous two-quantum processes in atoms can, under certain conditions, excite a resonant system much more effectively than the radiation of ordinary one-photon sources. This phenomenon was explained by the extremal strong quantum fluctuations of the intensity of the radiation, which are caused by the simultaneous emission of the two photons in two-quantum decays. The level of the intensity fluctuations in this case can be much higher than the mean intensity.

As a rule, classical fields from ordinary sources of broad-band radiation have an ordinary level of intensity fluctuations. Nevertheless the excitation of a resonance system by such a field can be made anomalously efficient without resorting to extremely sophisticated devices to strengthen the fluctuations.

It will be shown in the present paper that an increase of the excitation efficiency of the radiation can be produced by pairwise correlation of all the harmonics of its spectrum. The frequencies ω_0 and ω_1 of mutually correlated harmonics must be connected by the condition $\omega_0+\omega_1=\omega_g$, where ω_g is a frequency close to ω_{21}, such that $|\omega_g-\omega_{21}|\ll\Delta$. This possibility is demonstrated experimentally in the present paper with the example of the conversion of low-efficiency complex Gaussian noise into an efficiently acting correlated signal with nearly the same spectrum and intensity. The required correlation between the harmonics was accomplished by heterodyning of the original signal.

The role of correlation of the harmonics in the process of two-quantum excitation can be made clear as follows. The amplitude of the two-quantum transition is proportional to the product of the complex amplitudes a_0 and a_1 of the harmonics with frequencies ω_0 and ω_1. If there is no correlation between the harmonics, the phases of a_0 and a_1 are statistically independent and the ensemble average value is $\langle a_0a_1\rangle=0$. A correlation ensures that the average $\langle a_0a_1\rangle$ is not zero, as is also the case for a monochromatic field. Accordingly, it is formally to be expected that the action of radiation with pairwise correlation of the harmonics will have features of the action of a monochromatic field. This analysis is of course not exhaustive, primarily because it leaves aside the question of the role of interferences between different corresponding pairs, say ω_0, ω_1 and ω_0', ω_1' (see Fig. 1). The effect of the correlation between the harmonics on the two-quantum excitation characteristics can be explained in detail only in the framework of an adequate theory, one version of which is now to be expounded.

THEORY

Let us study the two-quantum excitation of a two-level system by nonmonochromatic radiation with the field strength

$$\mathcal{F}(t)=F(t)e^{-i\omega t}+F^*(t)e^{i\omega t}, \tag{1}$$

where $F(t)$ is a stationary random function of the time with a bell-shaped spectrum, whose width Δ is much smaller than the frequency ω_{21} of the transition, but at the same time is larger than all other parameters of the problem that have the dimensions of frequency. It is assumed that $\langle F(t)\rangle=0$. The other correlation characteristics of the field $F(t)$ will be indicated later. The excitation is close to resonance in the sense that ω is close to half of the transition frequency: $|2\omega-\omega_{21}|\ll\omega_{21}$. The actual value of ω is chosen so that the center of the spectrum of the signal $F(t)$ will be at zero frequency.

In the resonance approximation the dynamics of the two-level system in the field $F(t)$ is described by equations for the density matrix in the interaction representation:

$$\begin{aligned} i\dot\rho_{21}&=\varepsilon\rho_{21}+2W^2z,\\ i\dot\rho_{12}&=-\varepsilon^*\rho_{12}-2W^{*2}z,\\ i\dot z&=-i\gamma_1 z+W^{*2}\rho_{21}-W^2\rho_{12}+i\gamma_1/2. \end{aligned} \tag{2}$$

Here the following notations have been introduced: γ_1^{-1} is the lifetime of the excited state; $2z=\rho_{11}-\rho_{22}$; W^2 is the Rabi frequency of the two-quantum excitation, a complex quantity proportional to $F^2(t)$, $W^{*2}\propto F^{*2}(t)$; $\varepsilon=\omega_{21}-2\omega-i\gamma_2+\alpha WW^*$ is the complex resonance defect, in which expression the meaning of the first two terms is obvious, γ_2 is the phase relaxation frequency, and the last term determines the dynamic Stark (field) shift of the transition frequency and is a dimensionless real coefficient determined by the ratio between the field shift and the absolute value of the Rabi frequency.

The equations (2) describe the dynamics of the system in the rapidly varying fields $F(t)$ and $F^*(t)$. Our practical interest is in the quantities $\langle\rho_{21}\rangle$, $\langle\rho_{12}\rangle$, and $\langle z\rangle$, which are averages over the random realizations of the field. In the case when the width of the spectrum of the field that perturbs the system is much larger than all the frequencies characterizing the problem, the kinetic equations can be derived rigorously either by the method of functional derivatives, in the so-called diffusional-random-process approximation,[7] or by summation of the main sequence of the perturbation-theory series, which leads to the Dyson equation.[8] For simplicity in this exposition we shall give a formal derivation of the kinetic equation, decoupling the correlation between the dynamics of the system and the field fluctuations. The system of equations so derived agrees with the results of the rigorous methods, and it is this which justifies some lack of rigor in the derivation.

To shorten the formulas we introduce a column vector x with the components (ρ_{21},ρ_{12},z), the matrix

$$B=\begin{pmatrix} \varepsilon & 0 & 2W^2\\ 0 & -\varepsilon^* & -2W^{*2}\\ W^{*2} & -W^2 & -i\gamma_1 \end{pmatrix} \tag{3}$$

and a column vector q with the components $(0,0,\gamma_1/2)$.

We write the system (2) in this notation

$$i\dot{x}=Bx+iq \tag{4}$$

and average this equation over the ensemble of realizations of the field:

$$i\langle\dot{x}\rangle=\langle Bx\rangle+iq=\bar{B}\langle x\rangle+\langle\tilde{B}x\rangle+iq. \tag{5}$$

Here $\bar{B}\equiv\langle B\rangle$ is independent of the time, and $\tilde{B}\equiv B-\bar{B}$. It can be seen that x in Eq. (5) is determined by the quantity $\langle\tilde{B}x\rangle$; an equation for this quantity can be obtained in the following way. We integrate Eq. (4), then multiply by $\tilde{B}_t$ and average. The result is

$$\langle\tilde{B}_t x_t\rangle=-i\bar{B}\int^t\langle\tilde{B}_t x_{t'}\rangle dt'-i\int^t\langle\tilde{B}_t\tilde{B}_{t'}x_{t'}\rangle dt'. \tag{6}$$

Here the indices of all quantities refer to the time argument. The quantities $x_{t'}$ and $\tilde{B}_t$, taken at different times, are statistically independent, since x cannot follow the rapid variations of $\tilde{B}$. This fact makes it possible to uncouple the correlators and put Eq. (6) in the form

$$\langle\tilde{B}_t x_t\rangle=-i\int_{-\infty}^t\langle\tilde{B}_t\tilde{B}_{t'}\rangle\langle x_{t'}\rangle dt' \tag{7}$$

for all $t>-\infty$. Substituting (7) in Eq. (5), we get the desired kinetic equation

$$\langle\dot{x}_t\rangle=-i\bar{B}\langle x_t\rangle-\int_{-\infty}^t\langle\tilde{B}_t\tilde{B}_{t'}\rangle\langle x_{t'}\rangle dt'+q. \tag{8}$$

Hereafter we shall be interested only in the stationary state of the system. Its description is given by the solution of Eq. (8) for $t\to\infty$:

$$\bar{x}=(Q+i\bar{B})^{-1}q, \tag{9}$$

where $\bar{x}\equiv\langle x_{t\to\infty}\rangle$ and

$$Q=\int_0^\infty\langle\tilde{B}_t\tilde{B}_0\rangle dt. \tag{10}$$

The quantities $\bar{B}$ and Q that appear in Eq. (9) are the averages of binary and tetradic forms in the amplitudes F and F^*, and therefore to find the explicit form of the solution x we must take a specific form of the statistics of the field $\mathcal{F}(t)$.

Let us consider the case in which $W(t)$, a quantity proportional to $F(t)$, is a signal of the form

$$W(t)=V(t)\cos\theta+V^*(t)\sin\theta, \tag{11}$$

where $V(t)$ and $V^*(t)$ have the statistics of a complex Gaussian process, defined by the correlators

$$\langle V(t)V^*(t')\rangle=K(t-t'),\quad \langle V(t)V(t')\rangle=0, \tag{12}$$

and the coefficients $\cos\theta$ and $\sin\theta$ are introduced so that the quantity $\langle W(t)W^*(t)\rangle$, which is proportional to the intensity of the excitation, will remain unchanged for various relations between the terms in Eq. (11).

We point out an important difference between the statistics of the signal and those of the Gaussian signal $V(t)$. To do so we represent $W(t)$ in the spectral form

$$W(t)=\sum_\mu w_\mu e^{-i\mu t}=\sum_\mu(v_\mu\cos\theta+v_{-\mu}^*\sin\theta)e^{-i\mu t}. \tag{13}$$

As is well known, the different harmonics of the signal $V(t)$ are not correlated:

$$\langle v_\mu v_{\mu'}\rangle=0,\quad \langle v_\mu v_{\mu'}^*\rangle=q_\mu\delta_{\mu\mu'}, \tag{14}$$

whereas distinct, but adjacent harmonics of $W(t)$ are correlated:

$$\langle w_\mu w_{\mu'}\rangle=q_\mu\delta_{\mu,-\mu'}\sin 2\theta. \tag{15}$$

It is obvious that the angle θ characterizes the degree of correlation, and the statistics of the field are determined by two correlators:

$$\langle W(t)W^*(t')\rangle=K(t-t'),\quad \langle W(t)W(t')\rangle=K(t-t')\sin 2\theta, \tag{16}$$

the latter of which directly determines the values of the transition amplitudes in the dynamical matrix $\tilde{B}$. The relaxation matrix Q depends to a lesser degree on the correlation between adjacent harmonics.

Further transformations involved in calculating $\bar{B}$, Q, and $\bar{x}$ are obvious. Because of their cumbersomeness we omit them here. We present only the final expression for the stationary population $\bar{\rho}_{22}=\frac{1}{2}-\bar{x}$:

$$\bar{\rho}_{22}=\frac{1}{2}\left\{1+\frac{\gamma_1}{8\Gamma}\frac{(\omega_{21}-2\omega+\alpha K)^2+G'(G'+8\Gamma\sin^2 2\theta)}{(\omega_{21}-2\omega+\alpha K\cos^2 2\theta)^2+G(G+K^2\sin^2 2\theta/2\Gamma)}\right\}^{-1}. \tag{17}$$

Here we have shortened the formula by introducing the abbreviations $K=K(0)$, $G=\gamma_2+\Gamma(\alpha^2+4)\cos^2 2\theta$, $G'=G+2\Gamma\alpha^2\sin^2 2\theta$, and

$$\Gamma=\int_0^\infty K^2(t)\,dt. \tag{18}$$

The result (17) is a cumbersome expression with many parameters. It is difficult to analyze it in the general form. We confine ourselves here to the consideration of two limiting cases, completely uncorrelated ($\theta=0$) and maximally correlated ($\theta=\pi/4$) excitations. For the case with $\theta=0$

$$\bar{\rho}_{22}=4\Gamma/(8\Gamma+\gamma_1). \tag{19}$$

The first striking thing here is that $\bar{\rho}_{22}$ does not depend on ω. The absence of a frequency dependence of the population is a consequence of the insufficient accuracy of our theory, which does not describe effects associated with the shape and position of the center of the broadband excitation. In principle it is not difficult to perfect the theory so as to describe such effects, but we confine ourselves here to the results of the crude theory. As can be seen from Eq. (19), saturation in the case of uncorrelated excitation is characterized by an intensity that is determined by the relation $8\Gamma\approx\gamma_1$, or, if we allow for the fact that in order of magnitude $\Gamma\sim K^2/\Delta$ [see Eq. (18)], the condition for strong excitation takes the form $K^2>\gamma_1\Delta$.

Let us now discuss the case $\theta=\pi/4$, but first point out that Δ is assumed to be much larger than all characteristic frequencies of the problem, including the Rabi frequency $\sim K$, and therefore $\Gamma\ll K$. Furthermore we confine ourselves to the discussion of very low intensities, such that $K^2\ll\gamma_1\Delta$. At such intensities $\Gamma\ll\gamma_1\lesssim\gamma_2$, and therefore the uncorrelated field excites the system only weakly. Finally, assuming that α is not larger than a few times unity and neglecting small quantities of the order of γ_2/Δ and Γ/γ_2, we write the expression for $\bar{\rho}_{22}$ in this case:

$$\bar{\rho}_{22}=\frac{1}{2}\left\{1+\frac{\gamma_1}{8\Gamma}\frac{(\omega_{21}-2\omega+\alpha K)^2+\gamma_2^2}{(\omega_{21}-2\omega)^2+\gamma_2 K^2/2\Gamma}\right\}^{-1}. \quad (20)$$

In this expression we call attention to the dependence of $\bar{\rho}_{22}$ on ω. At first glance the very fact that $\bar{\rho}_{22}$ depends on ω disagrees with our earlier remark about the accuracy of the theory. The apparent contradiction is explained by the fact that in the case now considered ω has been given a double meaning. First, ω is the average frequency of the signal $\mathcal{F}(t)$, and secondly, it is at the same time the frequency relative to which the harmonics that have been correlated were paired off in Eq. (15). In assigning the same value to two frequencies that in general do not depend on each other, we avoided unnecessary complication in the theory. It can be shown that in the general case, when the two frequencies are different, the reaction of the system depends weakly on the first value and is essentially determined by the second value for correlated excitation.

Let us examine in detail the frequency dependence in Eq. (20). With the restrictions on the parameters that have been stipulated it is not hard to show that near $\omega\approx\omega_{21}/2+\alpha K/2$ the quantity $\bar{\rho}_{22}$ takes a maximum value:

$$\bar{\rho}_{22}=\frac{2K^2}{4K^2+\gamma_1\gamma_2}. \quad (21)$$

It is obvious that in this case saturation corresponds to intensities fixed by the condition $K^2\approx\gamma_1\gamma_2$, i.e., to values which characterize the saturation in a resonance monochromatic field,[9] and which are much smaller, by a factor $(\gamma_2/\Delta)^{1/2}$, than are necessary with an uncorrelated excitation.

In the neighborhood of the maximum the shape of the spectrum is approximately symmetric. Its width is characterized by the quantity $K(\gamma_2/\gamma_1)$, which determines the width of the resonance in the case of monochromatic excitation.[9] The parallel between the actions of monochromatic and correlated nonmonochromatic fields can be taken further. In a certain region of the parameters the population has a nonmonotonic dependence on the intensity of the correlated field. The picture is like that in the monochromatic case, the maximum being reached at an intensity given by the relation

$$K=-\frac{(\omega_{21}-2\omega)^2+\gamma_2^2}{\alpha(\omega_{21}-2\omega)}. \quad (22)$$

Specific features of nonmonochromatic excitation that appear in this case are as follows. As $\omega\to\infty$ the population does not go to zero, but comes to a level given by Eq. (19). This means that the spectral excitation contour consists of a narrow resonance on a broad background. The width of the latter is of the order of Δ. Besides this, the $\bar{\rho}_{22}(\omega)$ contour is not symmetrical. This asymmetry is nost marked in the neighborhood of values $(\gamma_2\Delta)^{1/2}$ and becomes stronger with increasing intensity and with decrease of the width Δ of the spectrum. This last effect, like the asymmetry of the line in the dynamic Stark effect,[2] is an example of the fact that a narrowing of the spectrum of an applied field can strengthen the difference between the system's behaviors in monochromatic and nonmonochromatic fields.

We now summarize the results of our theoretical examination of two-quantum excitation of a resonant system by a nonmonochromatic field. The theory predicts a strong effect of the degree of correlation between the harmonics in the spectrum of the applied field on the efficiency and other characteristics of the excitation. For the case of the correlation considered here this influence begins to show up at small values of θ, when [see Eq. (17)] $K^2\sin^2 2\theta/2\Gamma$ becomes larger than G. This means that a small admixture of correlated signal in Eq. (11) strongly changes the nature of the action of a nonmonochromatic field. The theory also predicts that the behavior of the system will be the same in correlated nonmonochromatic and in monochromatic fields. We have carried out experimentally a test of these conclusions and of the details of the theory.

EXPERIMENT

Two-photon excitation by nonmonochromatic radiation with controlled statistics was investigated experimentally with the Zeeman transition in the ground state of the Cd^{113} atom. The method of observation with the Cd^{113} oriented by optical pumping has been described in detail in Refs. 2 and 9; therefore we shall deal only briefly with the general scheme of the experiment.

The ground state $^1S_0$ of Cd^{113} has nuclear spin $\frac{1}{2}$ and splits in a constant magnetic field $H_z = 5G$ into two levels. The frequency of the transition between these levels is $\omega_{21}\approx 5$ kHz. The absence of other transitions in a wide range of frequencies near this value allows us to regard this atomic system as a two-level one as affected by applied fields. A predominant population of one of the Zeeman levels was produced by the method of optical orientation. To do this, Cd^{113} vapor was irradiated with circularly polarized resonance radiation at 3261 Å from a lamp filled with the isotope Cd^{114}. The population difference achieved in our experiments ($\sim 10^{-2}$) was sufficient for the observation of the absorption of radiofrequency quanta. The initial signal-to-noise ratio $\gtrsim 10^3$ made it possible to make experiments with intense fields, capable of saturating the atomic transition.

The arrangement of the experiment is shown in Fig. 2. A cell containing saturated Cd^{113} vapor at 220°C was placed at the center of a system of three pairs of Helmholtz coils, which produced a constant field H_z, a field $\mathbf{H}_0(t)=H_0(t)\{\mathbf{e}_x\cos[\omega_0 t+\varphi_0(t)]+\mathbf{e}_y\sin[\omega_0 t+\varphi_0(t)]\}$ rotating around the z axis, and a field $\mathbf{H}_1(t)=H_1(t)\mathbf{e}_z\cos[\omega_1 t+\varphi_1(t)]$ oscillating along the z axis.

Because of the selection rule $\Delta m = 1$ for the magnetic quantum number, two-quantum excitation is possible only through the combined action of the two fields $\mathbf{H}_0(t)$ and $\mathbf{H}_1(t)$.

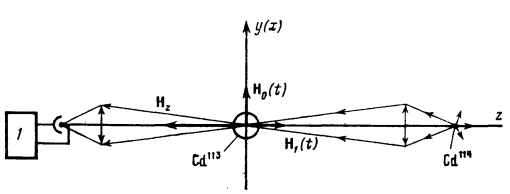

FIG. 2. Scheme of the experimental arrangement. 1—synchronous detector and automatic recorder.

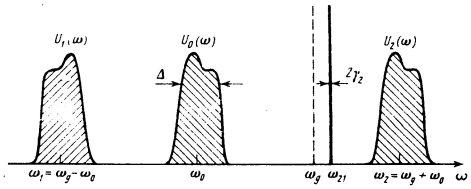

FIG. 3. Scheme for producing mutually complementary fields.

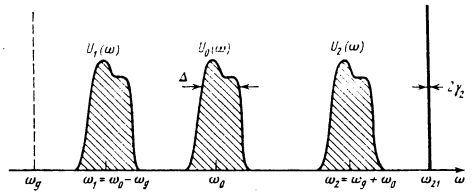

FIG. 5. Scheme for shaping fields in control experiments, in which the correlation gives no effect ($\omega_g < \omega_0$).

Let us consider in more detail the procedure for producing the frequency-phase correlation of the fields $H_0(t)$ and $H_1(t)$. Two limiting cases were realized in the experiment: 1) Maximal frequency-phase correlation of both fields, and 2) complete absence of correlation of both fields, although the method used to produce correlation, heterodyne transformation of the noise field with a monochromatic heterodyning signal, allows one in principle to realize intermediate situations.

The noise field $H_0(t)$ was produced by applying to the Helmholtz coils an RF voltage $U_0(t)$ produced by sending a broad-band, practically white, shot noise through narrow-band filters. It is well known that such a signal has the statistics of a complex Gaussian process.

The field $H_1(t)$ was produced with a noise RF voltage $U_1(t)$, obtained by nonlinear mixing of the noise $U_0(t)$ and a monochromatic heterodyning signal $U(t)$ and then filtering the resulting noise signal. In fact, the nonlinear mixing of the two signals $U_0(t)$ and $U_1(t)$ yields

$$U_0(t)U(t) = U_0' U \cos(\omega_0 t + \varphi(t)) \cos(\omega_g t + \varphi) = U_1' \cos(\omega_1 t + \varphi_1(t)) + U_2' \cos(\omega_2 t + \varphi_2(t)). \quad (23)$$

Accordingly, the heterodyning produces two signals with spectra centered near the average frequencies ω_1 and ω_2, and the spectral components of the original signal go over into the components of the new signals U_1 and U_2 in the following way: $U_0(\omega_0 - \delta\omega) \to U_1(\omega_1 + \delta\omega)$ and $U_0(\omega_0 + \delta\omega) \to U_2(\omega_2 + \delta\omega)$. Figure 3 shows the scheme of the nonlinear mixing process described by the expression (23).

Although the fields $H_0(t)$, $H_1(t)$, and $H_2(t)$ are mutually correlated, each one considered separately is a random function of the time with the statistics of a complex Gaussian process. The correlated nature of these fields becomes visible only when they act together, causing efficient two-quantum excitation of the system (Fig. 4). We point out here that the correlated nature of the noise fields manifests itself only in specific schemes of excitation. For example, in the scheme of Fig. 4a the fields $H_0(t)$ and $H_2(t)$ do not reveal their frequency-phase correlation, although its presence is guaranteed by the manner in which $H_2(t)$ was produced. The correlation appears in the excitation scheme shown in Fig. 4b. Similar remarks apply also to the field $H_1(t)$.

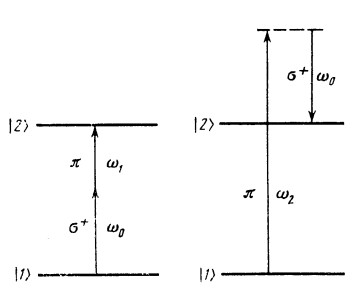

FIG. 4. Excitation scheme for two-photon transitions in which the correlation of fields manifests itself ($\omega_g > \omega_0$).

The reverse situation is also possible: With the same detection arrangement (Fig. 4), the frequency-phase correlation will be ruined if we change the way the fields $H_1(t)$ and $H_2(t)$ are produced. Figure 5 shows a different heterodyning scheme ($\omega_g < \omega_0$) which avoids the correlation of the fields $H_0(t)$ and $H_1(t)$ without shifting the average frequency $\omega_1 = \omega_0 - \omega_g$.

To allow a unique interpretation of the experiment in the cases of both correlated and uncorrelated fields we produced two-photon excitation of the atomic system only by the fields $H_0(t)$ and $H_1(t)$ with a transition according to the scheme in Fig. 4a. In this case the transformation according to the first scheme of Fig. 3 always assures correlation of the fields, and the second (Fig. 5) gives uncorrelated fields.

The effect of correlation of the exciting fields on the efficiency of the two-photon process can in principle appear with any spectral width of these fields. To make the effect very clear, however, and to show the possibility of strong excitation of the system by broad-band radiation, the width of the spectrum for the experiment was made large: $\Delta \gg \gamma_2$. This also fixes clean conditions for comparing the theory with the experiment. In fact, as has been shown in Ref. 2, at only moderate field intensities, when $\Delta \sim \gamma_2$, there is strong deformation of the excitation spectrum owing to dynamic shift of the levels, and the expected effect is masked.

The upper limit on the width of the spectrum, $\Delta \ll \omega_{21}$ has the obvious meaning of preventing one-photon excitation of the Zeeman transition by a wing of the spectrum of the noise field $H_0(t)$, since in our spin system one-photon transitions are also allowed by the selection rule. We chose the following two values for the width of the noise spectrum: $\Delta = 30$ Hz and $\Delta = 300$ Hz, whereas total width of the absorption line was $2\gamma_2 = 5.4$ Hz. These values fit into the indicated range and at the same time allow us to demonstrate the paradoxical narrowing of the excitation spectrum when the spectral width of the exciting radiation is increased.

It is simplest to investigate the population in two-photon excitation of a spin system by means of the one-photon absorption of a monochromatic probe field. Under conditions like these,[9] however, a nonlinear interference effect occurs which can complicate the interpretation of the observed phenomena. Therefore we chose a method

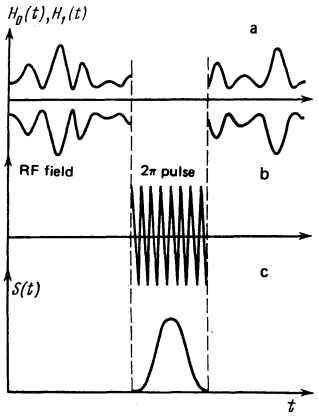

FIG. 6. Sequence of switching on of: (a) the noise fields $\mathbf{H}_0(t)$ and $\mathbf{H}_1(t)$, (b) the 2π pulse of registering field, and (c) the detection of the response $S(t)$ of the system to this pulse.

of observation which is free of side effects and consists of direct registration of the populations of the Zeeman levels. In our experiment this was achieved by direct-in the constant magnetic field H_z along the orienting optical beam (Fig. 2), which at the same time was the registering beam. With this scheme the observed signals are proportional to the deviation of the level population difference from the initial equilibrium value, and the maximum obtainable signals are proportional to the original value of the population difference.

To register the maximum signals, 2π pulses were applied to the atomic vapor with frequency 1 Hz. The noise The noise fields $\mathbf{H}_0(t)$ and $\mathbf{H}_1(t)$, by exciting two-photon transitions, diminished the population difference of the levels and thus also reduced the maximum signal corresponding to a 2π pulse. To lower the level of interferences, the noise RF fields were turned off during the passage of the 2π pulses. This procedure did not affect the amplitude of the signal, since the duration of a 2π pulse, $\tau = 5 \cdot 10^{-3}$ sec, was much shorter than the relaxation time $T = 0.09$ sec of the populations. The sequence of the applications of the fields is shown in Fig. 6.

The optical signal produced in a photoelectric element (Fig. 6) went to a synchronous detector and an automatic recorder. As the heterodyning frequency $\omega_g = \omega_0 + \omega_1$ was varied in the neighborhood of the transition frequency ω_{21} the value of the maximum signal (Fig. 6c) varied according to the law which governs the exitation spectrum of a spin system.

Figure 7 shows traces of the excitation spectra which are plots of the population difference $N = N_1 - N_2$ vs. the heterodyning frequency ω_g. In the comparison with the theoretical data we used not a curve averaged over several recordings, but a typical trace of the excitation spectrum at $K = \gamma/2$ and $\alpha = 2$. Here, according to the conditions of the experiment, $\gamma_1 = \gamma_2 = \gamma$. The quantity Δ was regarded as a fixed characteristic of the filter. As is seen in Fig. 7, the theoretical and experimental results are in good agreement.

The experiment demonstrates the possibility in principle of transforming complex Gaussian noise into a signal whose efficiency in two-quantum excitation of a

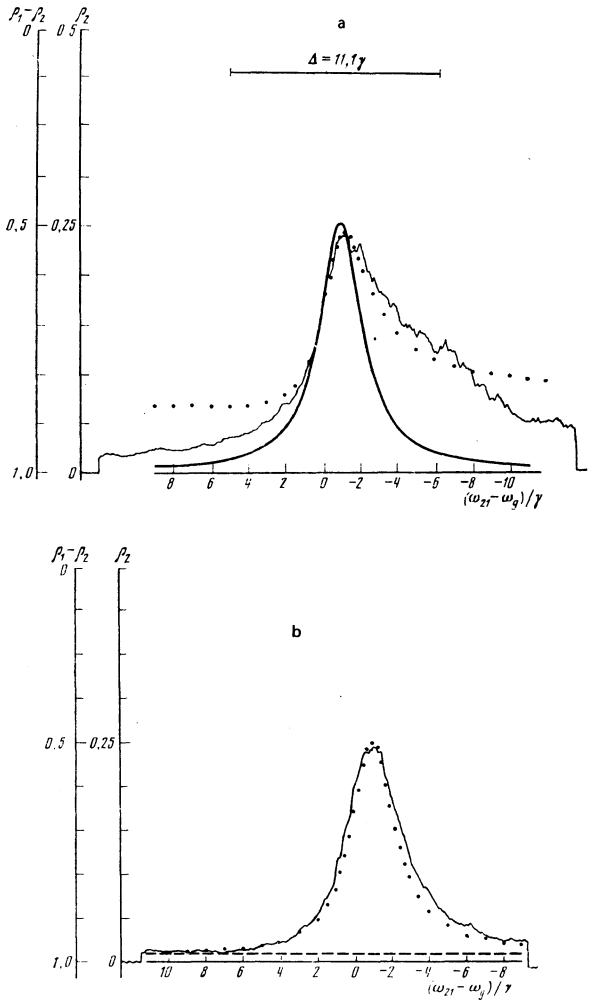

FIG. 7. Excitation spectra of the two-level system in mutually complementary fields at $K = \gamma/2$. a) $\Delta = 30$ Hz; b) $\Delta = 300$ Hz. The points are results of calculations with Eq. (17). The dashed line in Fig. 7b is the level of excitation without suitable correlation of the fields (see Fig. 5). The solid line in Fig. 7a is the excitation spectrum in a monochromatic field at the same power ($K = \gamma/2$).

resonant system is not inferior to that of a monochromatic field of the same intensity. It is well to emphasize once again that the method used here to produce the intraspectral correlation, namely heterodyning, makes no substantial change in either the spectrum or the level of intensity fluctuations of the field, as compared with the original field. At the same time the contrast between the effects of correlated and uncorrelated excitations are demonstrated not only by the shape of the spectra, as shown in Fig. 7, but also by the fact that with excitation by an uncorrelated field with $\Delta = 300$ Hz and with such a large intensity that $K \approx \gamma/2$, the population effect was about 10^{-2}, and under the same conditions a correlated field gave an effect $\approx \frac{1}{4}$ (Fig. 7b).

DISCUSSION OF THE RESULTS

Theoretical and experimental investigation has shown that the character and efficiency of the process of two-quantum excitation of a two-level system by a stochastic field can depend radically on the statistics of the field. The action of a correlated field has interesting

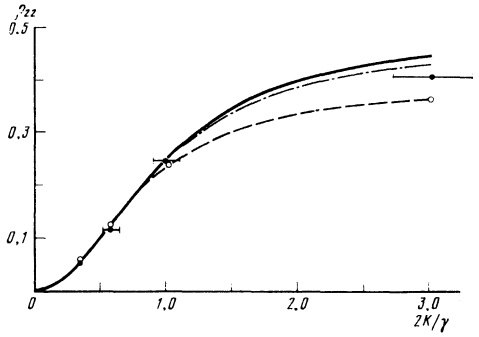

FIG. 8. Dependence of maximum population of upper level on $K \sim I^2$ (I is the intensity of the exciting field); the black circles are experimental with $\Delta = 300$ Hz; the dot-dash curve is theoretical for that case, and the solid curve is calculated for a monochromatic field; the open circles are experimental for $\Delta = 30$ Hz, and dashed curve is theoretical for the case. The horizontal bars correspond to the errors in determinations of the quantity K.

peculiarities, combining features of monochromatic and uncorrelated broad-band excitations. We shall single out and discuss some of these.

1. The spectrum of the excitation consists of a narrow resonance line and a broad background. The width of the latter is of the order of the width Δ of the spectrum of the stochastic field. The contrast between the background and the resonance line depends on the degree of correlation between the harmonics, which is determined by the value of θ. With broadband excitation the contrast is already significant with weak correlation ($\theta \ll 1$). With maximum correlation ($\theta = \pi/4$) the height of the resonance peak is the same as in a monochromatic field, which corresponds to the same excitation intensity as with a monochromatic field.

2. The width of the resonance line can be close to that found with monochromatic excitation of the same system. The resonance line is asymmetric, however, unlike that from monochromatic excitation. The degree of asymmetry increases with increasing intensity and with narrowing of the spectrum of the broadband radiation. This phenomenon is due to the dynamic shift of levels in stochastic fields, which has been discussed earlier[2] in connection with one-quantum excitation.

3. The pattern of excitation by a correlated field undergoes a paradoxical change, approaching that for a monochromatic field as the width Δ of the spectrum is increased. The change is accompanied by a narrowing of the resonance line in the spectrum of the excitation although also the contrast between the effects of correlated and uncorrelated fields increases with Δ (Fig. 8). This dependence of the shape of the excitation spectrum on the width is caused by the stochastic amplitude modulation which is always present in Gausian noise. The narrowing effect, like the asymmetry, appears also in one-photon excitation of resonant systems[2,8] and is due to the inertia of the field shift and splitting of levels. From the point of view of spectral representations the narrowing occurs because with broad spectra the interference due to the action of individual harmonics is suppressed. This enables us to understand the role of interference, mentioned earlier, in the process of two-quantum excitation of a resonant system by correlated broad-band radiation.

4. For a given sign of the detuning of the heterodyning frequency from the frequency for two-quantum resonance, the population becomes a nonmonotonic function of the intensity of the correlated field. This effect, like that produced with monochromatic radiation,[9] is due to competition between the population and the field shift of the levels. Although the nature of this phenomenon is the same for correlated and for monochromatic fields, the dependence of the population on the intensity can be very different. This fact, like other differences between the actions of correlated and monochromatic fields, can be formally associated with the appearance of the field parameter Γ in the case of nonmonochromatic excitation. Variation of this parameter determines the various patterns of excitation in a correlated field.

This paper deals with the case of the action of a correlated stochastic field formed from complex Gaussian noise by a heterodyning method. It must be pointed out that the statistics of the original signal may be arbitrary and not hinder the conversion of the signal by heterodyning into a signal with efficient two-quantum excitation of a resonant system. The heterodyning process produces a signal with phase shifts relative to the original signal. In a certain sense such a signal is reversed in time. When the original signal and the heterodyned signal act together their fluctuations balance each other and a regular component appears. It must be kept in mind that the correlated property produced in this way can be observed only with a certain way of having them act together.

In conclusion we wish to call attention to the fact that the radical dependence on the statistics of the radiation is not peculiar to two-quantum resonance processes only. This sort of property is possessed by both resonant and nonresonant processes with an even number of quanta. This fact, and the peculiar features of the action of correlated fields on a resonant system that are found in this present work allow us to think that the method of intraspectral correlation will find wide application in various problems of nonlinear optics and quantum radiophysics.

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