Coherent nonlinear phenomena in stochastic fields with frequency-phase correlation

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Two-photon excitation of a two-level system (Cd^{113}-atom vapor) by broadband radiation is experimentally investigated. It is established that the correlation between the harmonics of *t* noise-field spectrum can ensure an excitation effectiveness not inferior to that of a monochromatic field. The correlation was produced in experiment by a heterodyning method which made it possible to transform a broadband signal with the statistics of a complex Gaussian noise into a signal that excites resonantly the atomic system. Under the conditions of saturation of the two-photon transition, when an absorption spectrum of a weakly monochromatic field was recorded, a nonlinear interference effect was observed. This is evidence of the high degree of coherence of the action of a correlated noise pump in two-photon processes.

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The theory of stochastic processes shows convincingly that the character of the action of electromagnetic fields comes close to being monochromatic as the exciting-radiation spectrum becomes narrower. This conclusion is perfectly valid for one-photon processes and extends to an overwhelming majority of multiphoton ones.

At present there are two known types of deviation from this rule for multiphoton transitions. In the first case classical or quantum fluctuations of the exciting field intensity can lead under certain conditions to a more effective excitation by the noise than by the monochromatic field. An important factor is the low absolute excitation efficiency in conjunction with narrow-band pumping.[1–4] In the second case the frequency-phase correlation of the noise field of practically arbitrary spectral width and intensity ensures the same excitation efficiency as monochromatic action.[5] The method proposed in Ref. 5 makes it possible to surmount such a significant obstacle to high excitation efficiency as the finite width of the spectrum, i.e., the nonmonochromaticity of the exciting fields. The method consists of obtaining pairwise correlation of all the noise-pump harmonics, which should be connected for this purpose by the condition $\omega_1 + \omega_2 = \text{const}$. The correlation of the noise-spectrum harmonic is effected by mixing, in a nonlinear element, the initial noise field $E_0(t)$ with the monochromatic heterodyne field E_h. The noise fields $E_1(t)$ and $E_2(t)$ produced by the nonlinear mixing $E_h E_0(t)$ have spectra whose frequencies satisfy the conditions (Fig. 1):

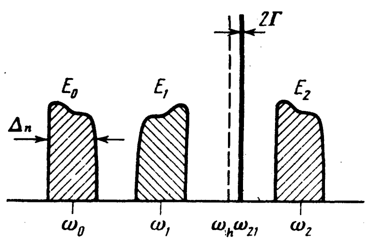

FIG. 1. Scheme for obtaining frequency-phase correlation of noise fields. Δ_n is the spectral width of the noise radiation.

$$\omega_0+\omega_1=\omega_h, \quad \omega_2-\omega_0=\omega_h.$$

The phases of the pairwise correlated harmonics satisfy similar equations. It must be emphasized here that the radiation statistics of the noise fields $E_1(t)$ and $E_2(t)$ do not differ from the statistics of the initial noise $E_0(t)$, and the intensity fluctuations of all the fields have the ordinary level.

The excitation spectra obtained in Ref. 5, which constitute the dependence of the level population difference on the heterodyne frequency, confirm so to speak the identity of the interaction of atomic systems both with correlated noise fields and with monochromatic ones. This has made it possible to suggest that nonlinear coherent effects can be observed under noise-pumping conditions.

One such effect, namely the nonlinear interference effect, consists in the appearance of regions where sounding radiation can be amplified by an atomic system without population inversion in the presence of a strong pump.[6] This effect was observed for both one-photon and two-photon monochromatic pumping.[7–9] In addition, it is observed under conditions of saturation of the atomic system by a nonmonochromatic field with a spectrum width on the order of the homogeneous width of the atomic line.[10]

In this paper are obtained spectra of two-photon absorption and a nonlinear interference effect under conditions when the atomic system is excited by stochastic fields with frequency-phase correlation.

EXPERIMENT

Two-photon noise excitation of a two-level system was observed on transitions between Zeeman sublevels of the ground state of optically oriented Cd^{113} atoms. The isotope Cd^{113} has in the ground state $^1S_0$ only a nuclear angular momentum 1/2, therefore in a constant magnetic field the population difference of the Zeeman sublevels was produced by optical orientation with a circularly polarized 3261 Å line emitted by an electrodeless quartz lamp with Cd^{112} (Ref. 7).

The experimental setup is shown in Fig. 2. The cell with cadmium vapor saturated at 220° C in an atmosphere of a

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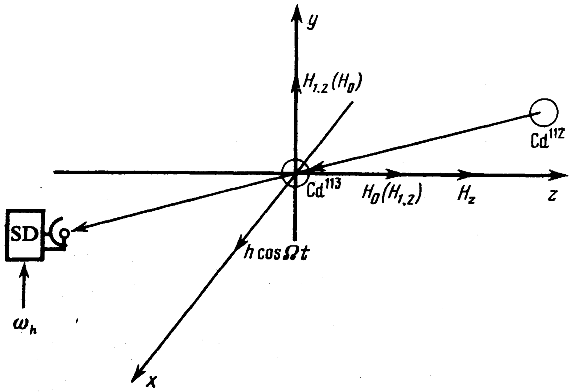

FIG. 2. Optical orientations of the Cd^{113} atoms. SD—synchronous detector.

buffer gas (nitrogen) was placed at the center of a system of three pairs of Helmholtz coils that produced mutually perpendicular fields: a dc field $H_z = 5$ G, which leads to the necessary Zeeman splitting of the ground state of cadmium with a frequency 5 kHz between the sublevels; radiofrequency fields H_0 and $H_{1,2}$, which participate in the two-photon excitation, and a sounding radiofrequency field $h \cos \Omega t$ at whose frequency the absorption was measured. The orienting light beam served simultaneously also for recording. In this study we recorded the modulation of the light-beam intensity at the motion frequencies of the transverse component of the atom polarization produced under the influence of the radiofrequency fields.The procedure described made it possible to obtain the absorption spectra of the two-level system at the frequency of any of the exciting fields.

The Zeeman sublevels of the ground state of cadmium can be regarded as a purely two-level system, since the frequencies of the optical transitions to the nearest levels are higher by several orders of magnitude than the frequency of the Zeeman splitting.

The two-photon excitation of the two-level system was effected by noise having complex Gaussian statistics. The high efficiency of the action of the noise radiation and its resonant character were ensured by pairwise correlation of the harmonics of the noise spectrum by the method described above (Fig. 1).

The conjugate noise fields produced by the heterodyning participated in the two-photon excitation in accord with the relations $\omega_0 + \omega_1 = \omega_h$ and $\omega_2 - \omega_0 = \omega_h$, in accordance with two schemes: the two-photon transition proper (Fig. 3a), and a transition of the type of Raman scattering (Fig. 3b). The effectiveness of the frequency-phase correlation manifested itself only in the specific two-photon excitation system (Fig. 3). Active in this experiment were either both mechanisms simultaneously or only one of them.

We investigated the action exerted on a two-level system by noise fields having spectral widths 300 Hz and 3 kHz at a resonance frequency 5 kHz and a natural line width 3.4 Hz. The noise fields were obtained from a white noise by using rectangular filters. Since the transition frequency was comparable with the noise-spectrum width, one-photon excitation by the wing of the noise field was possible in the system. To eliminate this effect, frequency rejection in the vicinity of the resonance frequency was used in the system that generated the radiofrequency fields.

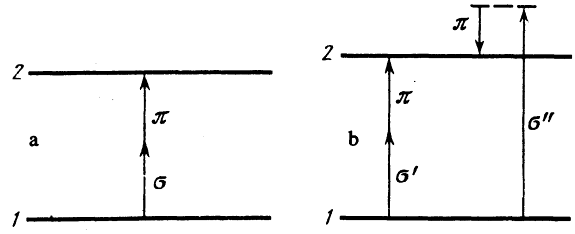

FIG. 3. Scheme of two-photon excitation of a two-level system.

DISCUSSION OF RESULTS

1. The two-photon absorption spectra of the correlated noise fields were obtained by detuning the central frequency $\bar{\omega}_{1,2}$ by an amount of the order of $\Gamma \ll \Delta_n$ (Γ is the natural line width and Δ_n is the spectral width of the noise radiation) and observing the absorption of the field $H_{1,2}$ at this frequency. Actually we monitored and retuned in the vicinity of the two-photon resonance the heterodyne frequency ω_h, which was used as the reference voltage (Fig. 2) when recording the spectra of the two-photon absorption (Fig. 4). It can be seen from Fig. 4 that they are narrow resonance lines with widths

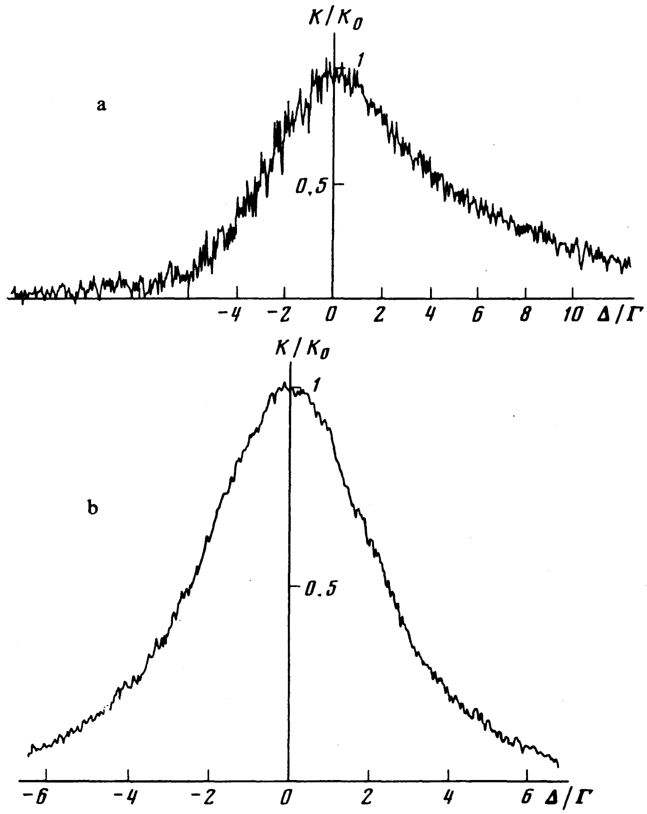

FIG. 4. Two-photon-absorption spectra of a correlated noise field: a—$\Delta_n = 300$ Hz; b—$\Delta_n = 3$ kHz. K—absorption coefficient of noise field, K_0—absorption coefficient of monochromatic field having the same intensity at the line center. Δ—frequency detuning of the field whose absorption is investigated from the transition frequency ω_{21}. Here $\Delta = \omega_0 + \omega_1 - \omega_{21}$.

much smaller than the width of the noise-pump spectrum. The spectra were normalized to the absorption coefficient of a monochromatic field under analogous conditions at the same field intensities.

The observed spectra differ little in practice from the two-photon absorption spectra in a monochromatic field. We note first the presence of an absorption-coefficient noise component due to stochasticity of the dynamic displacements of the levels in the pump field with fluctuating intensity, and to the nonstationary efficiency of the two-photon excitation. It can be seen from Fig. 4 that the level of the noise component increases as the noise-pump spectrum becomes narrower. Another difference, pertaining mainly to the absorption spectrum at $\Delta_n = 300$ Hz, consists in the appearance of an asymmetry of the absorption spectrum. A similar phenomenon was observed also in one-photon nonresonant action of a noise field on an atomic system.[10] The character of the asymmetry is connected with the insufficient degree of averaging of the stochastic instantaneous dynamic shifts of the levels during a time equal to the period of the average field-shift frequency.

A transition to the spectral width of the noise pump, equal to 3 kHz (the absorption spectrum is shown in Fig. 4b) leads to a considerable suppression of the noise component of the absorption coefficient, and in practice to a complete elimination of its asymmetry. The residual noise of the absorption coefficient is determined in the main not by the stochastic character of the excitation, but by the shot noise in the ~ 1 Hz recording band.

We have thus demonstrated in this part of the paper the use of the method of intraspectral correlation to produce a substantial change of the character of the interaction between noise and a resonant system. At noise-spectrum widths comparable with the transition frequency, the efficiency of two-photon excitation is indistinguishable from that in monochromatic fields (Fig. 4b), whereas for ordinary uncorrelated noise pumping the excitation efficiency is decreased compared with the monochromatic in a ratio Γ/Δ_n and has no resonant character. In our case, however, high saturations of the atomic transition become attainable at $\Gamma/\Delta_n \ll 1$, making possible observation of effects heretofore observed only in monochromatic or extremely narrow ($\Delta_n \approx \Gamma$) noise fields.[8,10]

2. The second and basic result of the described study was the observation of a nonlinear interference effect in a correlated noise. This effect, which is the consequence of the interference of the sounding and saturating fields in the course of the nonlinear interaction with the atomic system, turned out to be substantially dependent on the degree of averaging of the fluctuation component of the saturating field by the atomic system.

The absorption spectra of the sounding monochromatic radiation, for different widths of the exciting radiation and for different mechanisms of the two-photon excitation, are shown in Figs. 5a–5d. The experiments have shown that at a noise-spectrum width 300 Hz, i.e., much larger than the absorption line width, the stochastic character of the pump destroys the coherence of the interaction between the sounding and the saturating fields (Fig. 5a). The correlated character of the noise manifests itself here only in an appreciable atomic-transition saturation equal to the saturation in a monochromatic field of the same power, but experiment reveals strong fluctuations of the sounding-field absorption coefficient, and these mask the effect proper.

Thus, for noise pumping with spectral width 300 Hz, under conditions of frequency-phase correlation of the noise field, no nonlinear interference effect in the same form as in monochromatic pumping is observed. This effect consequently turned out to be more critical to the stochastic character of the exciting field than might be expected by starting from the two-photon absorption spectra (Fig. 4).

The fluctuations of the sounding-field absorption coefficient are considerably smaller at a noise-pump spectral width 3 kHz (Fig. 5c). Owing to the large spectral width of the noise, there is time for a considerable averaging of the instantaneous positions of the line center and of the fluctuations of the value of the field splitting of the levels, and regions where the absorption-coefficient sign is reversed appear in the absorption spectrum of the sounding field.

As already mentioned briefly above, the noise component of the absorption coefficient is due, first, to intensity fluctuations of the exciting-field component that leads to the stochastic field-induced level shift and to irregular, in time, deviations of the atomic systems from two-photon resonance with the saturating field, and second to the stochastic fluctuations of the probability of the two-photon transition. Whereas the transition-probability fluctuations cannot be eliminated in practice, the time-irregular field-induced shift of the levels can be suppressed to a considerable degree by using another scheme of two-photon excitation of the system (Fig. 3b).

According to the selection rules and to the principle of obtaining the frequency-phase correlation, there should participate simultaneously in the two-photon absorption the initial noise field H_0 and the conjugate fields H_1 and H_2, which appear in the heterodyning process and are located on both sides of the resonance line. In this case, when the initial noise field $H_0(t)$ was used as the circularly polarized component, a quadratic dynamic level shift, i.e., a resonance-line shift proportional to the noise-pump power, was observed in the system. On the other hand, when the rotating component responsible for the dynamic shift comprises simultaneously both transformed signals $H_1(t)$ and $H_2(t)$ (Figs. 5b and 5d), it becomes possible to eliminate this shift. Inasmuch as in this case the powers of the two fields were different and the frequency detunings of the centers of their spectra were of opposite sign and of equal magnitude, the field level shifts levels produced by them cancelled each other, causing an appreciable suppression of the stochastic fluctuations of the absorption coefficient. The sounding-field absorption spectrum acquires the structure typical of the nonlinear interference effect, and the maxima corresponding to transitions between quasi-energy sublevels become distinctly pronounced (Figs. 5b and 5d). It can be seen that the fluctuations of the absorption coefficient in monochromatic (Fig. 5e) and stochastic (Fig. 5d) excitation are comparable. For a com-

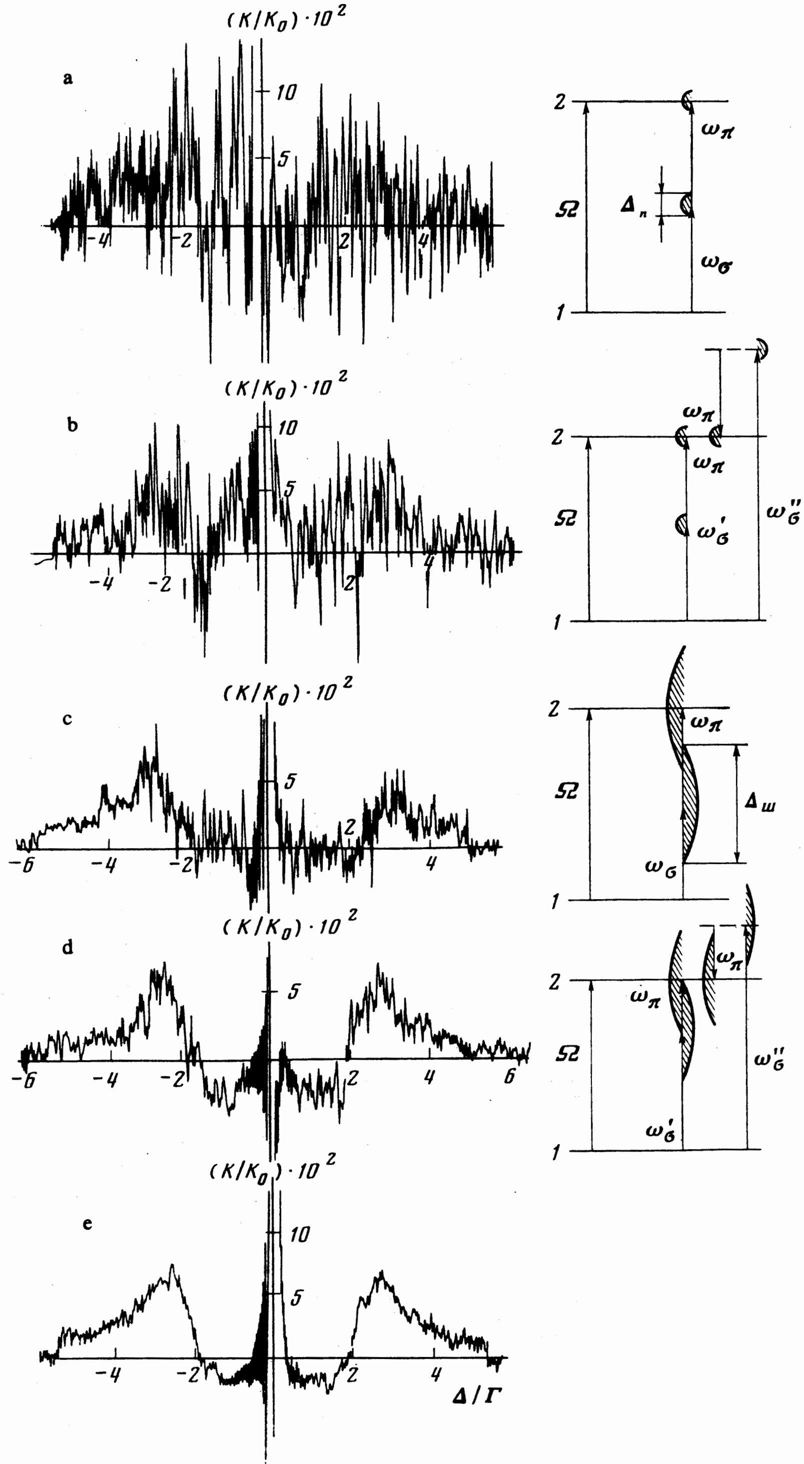

FIG. 5. Absorption spectra of weak sounding monochromatic radiation (nonlinear interference effect) for different widths of the noise-pump spectrum and different two-photon excitation mechanisms: a, b ($\Delta_n = 300$ Hz) and c, d ($\Delta_n = 3$ kHz)—noise pumping, e—monochromatic pumping. The shaded sections show the width Δ_n of the noise-field spectrum; $\Delta = \Omega - \omega_{21}$.

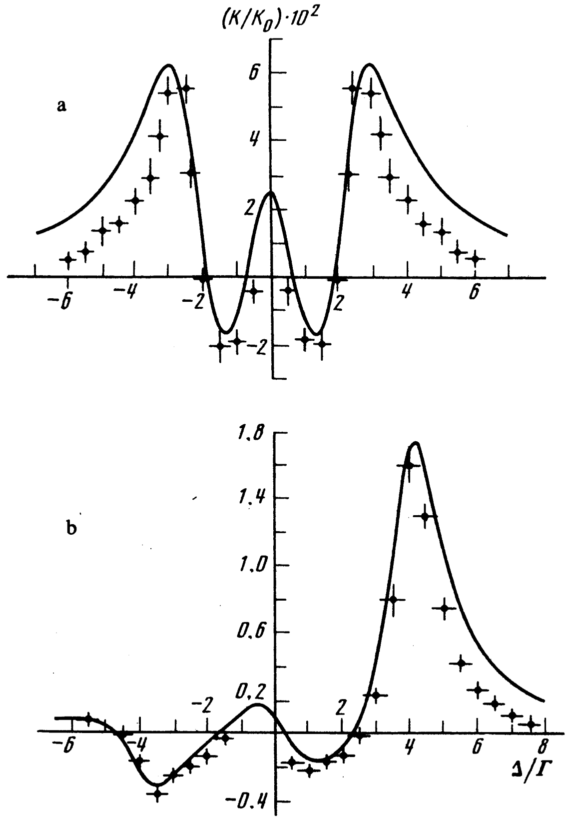

FIG. 6. Spectra of nonlinear interference effects: curves—theory, points—experiment ($\Delta = \Omega - \omega_{21}$): a—resonant two-photon action of saturating noise field, $\Delta_n = 3$ kHz, $V^2 = 5.25\,\Gamma^2$—saturation parameter, Γ—natural line width, $\varepsilon_0 = \omega_{21} - \omega_1 - \omega_0 = 0$; b—nonresonant action of noise field, $n = 3$ kHz, $V^2 = 14\,\Gamma^2$, $\varepsilon_0 = 1.4\,\Gamma$.

parison with the experimental curve we used calculations in accord with the equation for the sounding-radiation absorption coefficients in the case of two-photon monochromatic excitation.[8] Figure 6a shows the spectrum of the nonlinear interference effect in the case of resonant two-photon excitation of a strong noise field, while Fig. 6b shows the same for the nonresonant effect with $1.4\,\Gamma$ detuning of the saturating-field intensity from resonance. The agreement in the two cases is perfectly satisfactory. Thus, our investigations allow us to draw a number of conclusions.

First, the coherent nonlinear processes (as can be seen with the nonlinear interference effect as an example) turn out to be quite sensitive to the stochastic character of the radiation acting on the atomic system. An important role is played in this case both by frequency-phase and by amplitude fluctuations of the fields that participate in the nonlinear process. The elimination of the frequency and phase fluctuations, which lead to practically complete regularization of the two-photon absorption process, turned out to be insufficient to reveal subtle coherent effects of interaction of monochromatic and stochastic fields in a nonlinear medium. Amplitude fluctuations of the exciting radiation, which lead to stochastization of the field shift and to splittings of the atomic levels, average out to some degree because of the relaxation in the atomic system. On the other hand, the feasibility of eliminating the amplitude fluctuations by the method considered in the present paper calls for further research, as does also the question of applying this method to other types of radiation statistics.

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