An atom in the radiation field of a multifrequency laser

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This article analyzes the features of the interaction of a nonmonochromatic multifrequency laser radiation field with an isolated atom. Multiphoton excitation and nonlinear ionization of atoms under conditions in which the atomic levels are perturbed by the laser radiation field are treated. The extreme cases of interaction of an atom with a field are discussed in detail—the case of a broad laser radiation spectrum (fast field fluctuations) and the case of a narrow spectrum (slow fluctuations). The experimental data available in the scientific literature relevant to the set of questions under discussion are analyzed.

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CONTENTS

1. INTRODUCTION

A broad class of varied phenomena pertaining to nonlinear optics is based on various elementary nonlinear-optical phenomena that occur at the atomic level. As a typical example of such a phenomenon, we can point out the process of multiphoton excitation of an atom. In recent years the elementary nonlinear-optical phenomena have been studied experimentally in detail, many studies have been devoted to describing them theoretically, and the results of the studies have been generalized in a set of monographs.[1-4] As is well known, a sufficiently high intensity of the light field is required to allow observation of nonlinear phenomena. We must bear in mind that not only does the strong field give rise to multiphoton transitions in the atomic spectrum, but the very spectrum of the atom suffers substantial changes. Laser radiation is employed in experiments in practically all cases involving a strong light field. However, as a rule, the experimental data and the results of calculations pertain to external fields having substantially different properties. While most calculations have assumed the field to be strictly monochromatic, most of the experiments have been performed with nonmonochromatic laser radiation. This review will discuss the elementary nonlinear-optical phenomena that arise in the interaction of nonmonochromatic radiation in the visible frequency range with an isolated atom. Although the degree of monochromaticity $\Delta\omega/\omega$ of laser radiation is very high in comparison with the radiation of incoherent light sources, yet the width $\Delta\omega$ of the laser radiation spectrum is always finite. In many cases it is not at all small in comparison, e.g., with the width of the atomic levels, while the intensity F of the radiation fluctuates in time. The consequences of the nonmonochromaticity of laser radiation are nonlinear interaction characteristics differing from the case of ideal, strictly monochromatic radiation. A different absolute value of the probability is realized, as well as different dependences of the probability on the frequency and intensity of the radiation. The differences between the result of action of monochromatic and nonmonochromatic fields in exciting nonlinear optical phenomena can be most easily be described in the special case in which the nonmonochromatic field is a Gaussian random quantity. Although, strictly speaking, the nonmonochromatic laser radiation field is not Gaussian, yet it can be treated as such to a certain degree of accuracy. This situation is very important, since it simplifies the analysis of various phenomena.

In writing this review, we have eschewed the aim of presenting the entire variety of different phenomena that can arise in the nonlinear interaction of nonmonochromatic laser radiation with various objects at the atomic level. We wished to discuss only the fundamental phenomena that are most essential in drawing an overall picture of the interaction, and also essential in practice. Correspondingly, as a rule, an atom serves as the object, while the radiation field of a multifrequency laser serves as the nonmonochromatic

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field.

A laser is generally said to be a multifrequency one when it emits many longitudinal modes having the lowest transverse index. The radiation of a multifrequency laser is a typical example of nonmonochromatic radiation. Whenever the modes are not mutually coupled, the radiation field of a multifrequency laser is random. Both the number of generated modes and the character of the mutual coupling between them determine the averaged characteristics of the laser radiation: the width $\Delta\omega$ and the form $F(\omega)$ of the spectrum, the intensity distribution $P(F)$, and the higher-order correlation functions. An important feature of multifrequency laser radiation is the presence of temporal fluctuations of the emission intensity, so that the radiation field intensity proves to be a random quantity that can be described in the language of correlation functions.[5] The time scale of the fluctuations of the radiation field is determined by the correlation time $\tau_{cor} \sim 1/\Delta\omega$, i.e., the reciprocal of the spectral width of the radiation. Since the widths of the radiation spectra of multifrequency lasers lie in the range 0.1–10 cm^{-1}, the correlation times lie in the range 10^{-10}–10^{-12} s. For multifrequency laser radiation, the quantity $F(t)$ varies weakly within a time interval $\Delta t \leq \tau_{cor}$, while in a time interval $\Delta t \gg \tau_{cor}$, the quantity $F(t)$ fluctuates and takes on all possible values to realize the distribution $P(F)$.

In describing the nonlinear interaction of multifrequency laser radiation with an atom, one must establish which field it is that leads to the observed effect—the instantaneous value of the field or the value of the field averaged over some time interval? There is no unambiguous answer to this question. In various special cases the answer is determined by the relationship between the correlation time of the radiation and the response time of the atom (we assume that the observation time is always longer than the response time of the atom). In the case of an instantaneous response, when we can consider the fluctuation of the intensity to be slow (τ_{cor} large) the interaction with the nonmonochromatic radiation is equivalent to the interaction with monochromatic radiation having a slowly varying intensity (as compared with the response time). Here the width of the radiation spectrum plays no role, so that the yield of the nonlinear process is determined only by the distribution $P(F)$. Below we shall call this the case of a narrow radiation spectrum. In the case of a noninstantaneous response, in which the intensity fluctuates rapidly in time (τ_{cor} small), the character of the time variation of the field becomes important. Then the correlation functions of the field of increasingly higher order must be manifested in the probabilities of the nonlinear processes with increasing degree of nonlinearity of the process. Thus a dependence of the yield of a nonlinear process on the width and form of the radiation spectrum arises. We shall call this the case of a broad radiation spectrum.

We can point out three observable characteristics of nonlinear optical phenomena that are governed by the properties of the nonmonochromatic radiation. They are the mean yield of the nonlinear process over the time interval of observation, the fluctuations of the yield of the nonlinear process about the mean, and the dependence of the yield on the frequency of the radiation and the form of its spectrum. Although the listed characteristics are of equal importance, both from the standpoint of their role in the nonlinear interaction and from the standpoint of their mutual relation to the concrete properties of the radiation, we shall mainly discuss the mean yield and its dependence on the radiation spectrum, since the problem of fluctuations in the nonlinear interaction of laser radiation with atoms has been studied in insufficient detail.

The problem of the nonlinear interaction of nonmonochromatic radiation with an atom is often treated in two counterposed aspects. The point is that currently both the properties of the radiation of multifrequency lasers and the elementary nonlinear-optical phenomena *per se* are objects of study. Therefore nonlinear phenomena are sometimes viewed as a detector for studying the properties of the radiation, and sometimes radiation having some specified properties is employed for studying nonlinear phenomena. Both approaches to the problem under consideration will be closely interwoven below.

2. FUNDAMENTAL PROPERTIES OF MULTIFREQUENCY LASER RADIATION

As we have said above, we are considering as nonmonochromatic laser radiation the radiation that arises in a multifrequency generation regime of lasers that operate in pulsed and continuous-wave regimes. Multifrequency laser radiation is characterized by fluctuational intensity excursions in time that alternate with deep gaps (almost to zero). Correspondingly, multifrequency laser radiation must be characterized by the intensity distribution $P(F)$, the width $\Delta\omega$ and form $F(\omega)$ of the spectrum of the radiation, and also by the corresponding correlation functions. In principle, in a pulsed generation regime one should also take account of the finite pulse duration. However, in practice it does not contribute to the width of the multifrequency radiation spectrum.

Multifrequency laser radiation can be described in the language of the time variation of the amplitude $A(t)$ and the phase $\varphi(t)$ of the electric field intensity $\mathscr{E}(t)$:

$$\mathscr{E}(t) = A(t)\, e^{i\omega_0 t + i\varphi(t)} \qquad (2.1)$$

and of the radiation intensity $F(t) = \mathscr{E}(t)\mathscr{E}^*(t) = A^2(t)$ [in Eq. (2.1), ω_0 is the central frequency of the radiation spectrum of the laser].

One can also use the language of the mode composition of the radiation. In this language, the radiation amounts to the sum of the fields corresponding to the established intrinsic oscillations in the laser resonator (modes) at the different frequencies:

$$\mathscr{E}(t) = \sum_n A_n e^{i\omega_n t + i\varphi_n}.$$

Here A_n, ω_n, and φ_n are respectively the amplitude, frequency, and phase of the nth mode. The case in which we are interested concerns the longitudinal modes, whose frequencies form an equidistant sequence.

An obvious condition that allows us to employ the mode language is that the character of the radiation is stabilized. This arises when the duration of emission considerably exceeds the time of passage of the light through the laser resonator (the so-called axial period). The evident advantage of the mode language for describing multifrequency laser radiation consists of the fact that the amplitudes and phases of the modes are either constant, or are slowly varying functions of the time as compared with the duration of the excursions. The excursions themselves result from beating of the modes differing in frequency. Here the sequence of fluctuational excursions repeats in each axial period, while the number of excursions per axial period, i.e., the number of statistically independent values of the radiation intensity, is approximately equal to the number of modes being generated.

When one is employing multifrequency laser radiation to study nonlinear optical phenomena, the optical radiation is that whose fluctuations are described by a Gaussian random process. A well-known example of such radiation is the radiation of a thermal source. In this case the intensity distribution of the radiation is described by the exponential function

$$P(F)=\frac{1}{F_0}\exp\left(-\frac{F}{F_0}\right). \qquad (2.2)$$

The correlation functions of higher orders are expressed in terms of the first-order correlation function, which is the Fourier image of the radiation spectrum $F(\omega)$. The interpretation of the experimental data is most unambiguous when one employs such a laser radiation, while it suffices to know the mean intensity $\langle F\rangle = F_0$ and the form of the spectrum $F(\omega)$ in order to characterize the radiation.

The possibility of obtaining from a laser radiation having properties resembling those of a thermal source stems from the fact that laser radiation evolves from spontaneous noise. Evidently a regime of generating many modes is optimal for imitating the properties of the radiation of a thermal source. That is, the radiation spectrum must be sufficiently broad. Only under these conditions can the distribution $P(F)$ be close to the exponential distribution of (2.2). The width of the spectrum of the laser radiation, i.e., the number of generated modes, is determined by the properties of the active medium, the design of the laser resonator, and the generation regime. The distribution $P(F)$ for laser radiation can differ from exponential, not only owing to an insufficient number of generated modes, but also to nonlinear effects that arise in the laser resonator and which are manifested as coupling of modes.

First let us turn to the phenomenon of mutual coupling of modes. Many concrete factors are known that interfere with the independence of the generated modes. All the factors are based on the nonlinear interaction of the radiation with the material of the resonator. The appearance of these phenomena, together with their concrete influence on the mutual coupling of modes, substantially depends on the design of the laser, the particular fabrication of the standard design elements, and the operation regime of the laser. Examples of the appearance of a nonlinear interaction in the active medium of a laser resonator are well known, both from direct measurements of the metrics of the radiation,[6] and from the results of measurements performed with multiphoton detectors.[7] The nonlinear interactions have one common property: their role increases nonlinearly with increasing intensity of the radiation in the resonator. The effect of the nonlinear interaction on the quantity $P(F)$ is qualitatively clear. The nonlinear phenomena that curb the most intense fluctuation excursions limit the region of intensities in which $P(F)$ differs from zero. Automodulation and stimulated scattering possess this character. The effect of curbing of intense excursions is most clearly demonstrated by a model experiment[8] in which the laser radiation was subjected to the action of a nonlinear medium and then detected with a two-photon detector. The nonlinear phenomena that increase the amplitudes of the most intense fluctuational excursions expand the region of intensities in which $P(F)$ differs from zero. A well-known example is the bleaching of a nonlinear medium placed in a resonator under the action of the radiation field in the resonator. This effect has been observed by using the method of a multiphoton detector, with Q-switching of a neodymium-glass laser by means of a bleaching filter.[9] The essential point is that the nonlinear effects can arise directly in the active medium of the resonator.

The aggregate of experimental and theoretical data on nonlinear interaction in the active medium of a laser resonator allows us at present to realize a generation regime in which the modes of the laser radiation are independent. In practice, to do this, one must not exceed a certain field-intensity level in the resonator.

In contrast to the role of nonlinearity, the role of the number of modes can be taken into account in the form of rather general relationships. For continuous-wave laser radiation, the specific property that qualitatively distinguishes it from the radiation of a thermal source is the constancy of the energy emitted during the time of an axial period. The constancy of this energy stems from its strict correspondence to the number of active particles in the laser resonator, which is held constant in time.

We recall that the form of the fluctuations in one axial period in a continuous-wave generation regime differs somewhat from the form of the fluctuations in an adjacent period. Upon accumulating, these differences are expressed in the fact that the amplitudes and phases of the modes slowly vary in time (as compared with the axial period). However, in spite of these changes, the fundamental statement—the constancy of the energy emitted per axial period, remains in force. The energy emitted per axial period equals the sum of the energies of all the fluctuational pulses. Therefore the energy of the maximal fluctuational pulse cannot in any case exceed the energy per axial period. Consequently, the radiation intensity in a maximal excursion cannot exceed the mean intensity of the radiation per axial period by a factor of more than T/τ_{cor} (here T is the length of an axial period, and τ_{cor} characterizes the duration of

the fluctuational excursions). Thus the distribution $P(F)$ for continuous-wave laser radiation always differs qualitatively from the distribution (2.2) for a thermal source, in which the amplitude of a maximal excursion is not bounded, and the maximal intensity in an excursion can exceed the mean intensity by an arbitrary factor. Since the number of fluctuational excursions per axial period is unambiguously associated with the number of generated modes, we can establish the relationship of the distribution $P(F)$ to the number N of generated modes. The derivation of this relationship, which is given below, is based on the constancy of the energy emitted per axial period. The dependence of $P(F)$ on the number of generated modes is described by the relationship:

$$P(F)=\frac{N-1}{N\langle F\rangle}\left(1-\frac{F}{N\langle F\rangle}\right)^{N-2}, \qquad 0\leqslant F\leqslant N\langle F\rangle. \tag{2.3}$$

As $N\to\infty$, this goes over into the exponential distribution of (2.2) for the radiation of a thermal source. The relationship (2.3) has been derived in Refs. 10 and 11 under the assumption of a very simple, rectangular form of spectrum. It is important to note that differences between the distributions (2.3) and (2.2) exist throughout the interval of variation of F. We can see this well from Fig. 1, which shows the $P(F)$ relationship for different numbers of modes. Naturally these differences increase with decreasing number of modes.

The actual form of the radiation spectrum of a multifrequency laser differs from rectangular, being close to a Gaussian curve. An account taken of the Gaussian form of the spectrum, which has been carried out numerically in Refs. 12 and 13, has shown that Eq. (2.3) qualitatively describes correctly the intensity distribution, while it can be used quantitatively with sufficient accuracy if we employ in Eq. (2.3) the number of modes corresponding to twice the width of the Gaussian distribution.

Thus the distribution $P(F)$ is close to exponential when there is a large enough number of modes, and in practice we can neglect the dependence of $P(F)$ on the number of modes and on the form of the laser radiation spectrum. We note that the criterion of closeness of the actual distribution $P(F)$ to the exponential distribution of (2.2) is determined by the degree of nonlinearity of the process excited by the laser radiation. This criterion is given in Sec. 6.a.

Now we shall turn to the features of the pulsed regime

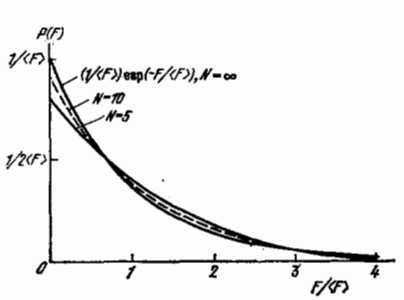

FIG. 1. Form of the distribution $P(F)$ of the radiation intensity of a multifrequency laser with varying numbers of generated modes $N=5,\ 10,\ \infty$.

of multifrequency generation of laser radiation.

The pulsed regime of generation, which can be realized by Q-switching of the laser resonator, amounts to a sequence of pulses the intervals between which exceed their duration by many orders of magnitude. A radiation pulse of a multifrequency laser contains a considerable number of fluctuational excursions. When one records the laser radiation with a detector whose time resolution is larger than the duration of the fluctuations, but smaller than the duration of the pulse, one can measure the time variation of the mean intensity $\langle F\rangle$, or the envelope of the pulse. The envelope of an individual pulse has the shape of a smooth, bell-shaped curve with fronts approximately equal to the duration of the pulse. As a rule, the latter lies in the range from several units to several tens of nanoseconds.

Since the radiation pulses are relatively rare, and we are measuring the result of the action of an individual pulse on the material, we are interested in the properties of the radiation in an individual pulse. The radiation in an individual pulse is characterized by practically invariant phases of the generated modes, and also by an invariant relationship between the amplitudes of the modes. As a rule, the energies of the individual modes differ substantially, reproducing the spectrum of the radiation only in the mean. When the phases and the relationships between the amplitudes of the generated modes are invariant, the distribution $P(F)$ does not vary functionally during a pulse, but only the mean intensity of the radiation varies. In a series of successive pulses, the values of the amplitudes and phases of the modes take on random values.

The energy in a pulse is bounded by the same factors as the energy emitted per axial period in a continuous-wave regime of generation, and also by the fact that the process of Q-switching of the resonator is reproduced in a sequence of successive pulses with sufficient accuracy. Thus the ideas expressed above on the nature of the distribution $P(F)$ hold also for a pulsed regime. Equation (2.3), which relates $P(F)$ to the number N of generated modes, also holds. Here, just as in the case of a continuous-wave emission regime, the quantity $\langle F\rangle$ must be measured in an interval of time exceeding the axial period.

Although the pulsed regime is a very widespread generation regime, and it is precisely in this regime that most of the data have been obtained on the nonlinear interaction of laser radiation with matter, yet due attention has not been paid to the model of pulsed radiation described above, in which not only the phases, but also the amplitudes of the modes are random. A model that assumes a deterministic character of the amplitudes of the modes has been employed in a set of studies[14,15] to describe laser radiation. We note that it allows one to draw only individual qualitative conclusions on pulsed radiation, and it does not give a correct quantitative answer to a number of problems.

We must also note that the widely employed model of laser radiation with a randomly "diffusing" phase is not applicable for describing multifrequency laser radia-

tion. This model reflects correctly the fundamental features of continuous-wave single-frequency radiation, for which the intensity is practically constant, while the phase is a random quantity. In this case the main effect that gives rise to the width of the spectrum is the variation of the phase. As we have stated above, in a multifrequency generation regime, the intensity of the radiation fluctuates substantially. Here the width of the radiation spectrum is primarily determined by the large number of generated modes, rather than by their phase variation.

3. POWER-FUNCTION PROCESSES IN A NONMONOCHROMATIC FIELD

This section will examine one of the very simple cases in which the nonlinearity of the studied phenomena arises solely from the multiphoton character of the transition of the electron from the initial to the final state, and the field intensity is substantially less than the atomic field intensity, while we can neglect the perturbation of these states by the exciting field. Thus we have restricted the treatment to power-function processes for which the probability in a monochromatic field is associated with the intensity of the radiation by the power-function relationship:

$$W \sim F^k.$$

Here k is the number of photons that must be absorbed to satisfy the law of conservation of energy. Very simple examples of such processes are the direct multiphoton ionization of atoms and the multiphoton excitation of atoms.

The multiphoton ionization of atoms in a field of frequency ω and intensity $\mathscr{E}$ is direct in character if we can neglect the probability of resonance transitions in comparison with that of nonresonance transitions, while the Keldysh adiabaticity parameter is $\gamma=\omega/\omega_{tun}=\omega\sqrt{2E_n}/\mathscr{E}\gg 1$, where ω_{tun} is the frequency of tunneling, and E_n is the binding energy of the electron.[16] In practice the direct process occurs in the visible frequency range at a field intensity $\mathscr{E}<\mathscr{E}_{at}$ and with a detuning of the intermediate resonances that substantially exceeds the width of the resonance states.

The probability of the direct ionization process in a monochromatic field is described by the relationship:

$$W_m(F)=\alpha_k(\omega)F^k. \tag{3.1}$$

Here $\alpha_k(\omega)$ is the multiphoton cross-section of this process. The multiphoton cross-section $\alpha_k(\omega)$ of the direct ionization process far from resonances is a slowly varying function of the frequency of the radiation. We shall neglect the variation of the cross-section as the frequency varies within the limits of the width of the radiation spectrum of the multifrequency laser. Therefore the width of the radiation spectrum plays no role in the direct process of multiphoton ionization. Correspondingly, the difference between ionization in a monochromatic and a nonmonochromatic field consists of the fact that we can assume the intensity to be constant in the former case, but to be a random quantity described by the distribution $P(F)$ in the latter case. The probability of ionization in a nonmonochromatic field is described by the obvious relationship:

$$W(\langle F\rangle)=\int_0^\infty W_m(F)P(F)\,dF. \tag{3.2}$$

Here $W_m(F)$ is the probability of ionization in a monochromatic field of intensity F, while $\langle F\rangle$ is the mean intensity of the nonmonochromatic radiation, which is a parameter of the distribution $P(F)$. In line with (3.1), the probability is:

$$W(\langle F\rangle)=\alpha_k\int_0^\infty F^kP(F)\,dF=\alpha_k\langle F^k\rangle. \tag{3.3}$$

Here the angle brackets denote averaging of the yield of this process over the ensemble of values of the radiation intensity that are realized in the time of measurement, while $\langle F^k\rangle$ is the kth moment of the intensity distribution $P(F)$. The standard problem arises: by starting with the measured values of the ionization probability in a nonmonochromatic field, to derive data on the probability in a monochromatic field. We note that the role of nonmonochromaticity becomes more substantial with increasing degree of nonlinearity (multiphoton character) k of the ionization process.

The accepted manner of taking into account the role of nonmonochromaticity is by employing the so-called statistical factor. When the frequency of the monochromatic radiation equals the central frequency of the nonmonochromatic radiation and the intensities of the two radiations are equal ($F_m=\langle F\rangle$), the statistical factor is commonly defined by the quantity:

$$g_k=\frac{W}{W_m}=\frac{1}{F_m^k}\int_0^\infty F^kP(F)\,dF. \tag{3.4}$$

In the case that the nonmonochromatic radiation is radiation from a thermal source and accordingly $P(F)$ is described by the exponential relationship (2.2), we can easily see from (3.4) that the statistical factor has the well-known value[1]:

$$g_k=k! \tag{3.5}$$

The statistical factor has the same value for laser radiation if the latter is equivalent to the radiation from a thermal source.

The relationships derived above, which describe multiphoton bound-free transitions in a nonmonochromatic field, break down if the ionization process begins to acquire a tunneling character with increasing radiation intensity (cf. Sec. 6, b), intermediate resonances arise (cf. Sec. 6, c), or if the concept of the transition probability per unit time proves inapplicable, owing to a long pulse duration of the exciting field.

Now let us turn to the case of multiphoton excitation of an atom. In contrast to bound-free transitions, bound-bound transitions exhibit a multiphoton cross-section of the transition that substantially depends on the frequency in the resonance region. Therefore the width of the spectrum of the laser radiation affects the transition probability. However, in the case that we have been treating, the character of this dependence does not vary with varying field intensity, since we assume that the perturbation of the resonance level is smaller than its natural width (the converse situation in

which the perturbation of the resonance state plays a substantial role is treated in Sec. 5).

A dependence of the cross-section on the frequency of the radiation in multiphoton excitation has the result that the kth moment of the intensity distribution does not suffice for characterizing the radiation, but we need a knowledge of the kth-order correlation function. In the absence of intermediate resonances, when the multiphoton transition amounts to a series of virtual single-photon transitions, the probability of the multiphoton transition is determined by the kth-order simultaneous correlation function[1,17]:

$$G_k(t) = \langle [\mathscr{E}^*(t')\,\mathscr{E}(t'+t)]^k \rangle. \tag{3.6}$$

Qualitatively the function $G_k(t)$ behaves in the same way as $G_1(t)$. When $t=0$, the value of the correlation function equals the kth-order moment of the intensity distribution $P(F)$:

$$G_k(0) = \langle F^k \rangle. \tag{3.7}$$

As t increases from zero, the function $G_k(t)$ substantially declines from the value $\langle F^k \rangle$ within the correlation time interval $\tau_k < \tau_1 \equiv \tau_{cor}$. In this interval $G_k(t)$ is modulated at the frequency $k\omega_0$, where ω_0 is the central frequency of the emission spectrum. The averaging in Eq. (3.6) is performed over the large number of independent values of $\mathscr{E}(t)$. Since in the generation regimes that we have examined (continuous-wave and pulsed regimes of generating axial modes) the mode composition of the radiation does not change, the choice of the instant of time t' plays no role. In this sense we can consider the laser radiation field to be stationary.

The probability of a k-photon bound-bound transition per unit time is described by the following relationship[1,17]:

$$W = A_k \int_{-\infty}^{\infty} G_k(t)\, e^{-i\omega_{01}t - \Gamma|t|}\, dt. \tag{3.8}$$

Here A_k is the compound matrix element of the transition, ω_{01} is the frequency of the transition between the states 0 and 1, and Γ is the natural width of the excited state, which is introduced phenomenologically.

As Eqs. (2.1), (3.6), and (3.8) imply, the frequency-dependence of the transition probability $W(\omega)$ is determined both by the lifetime of the atom in the excited state and by the decay time of the correlation function $G_k(t)$. The lesser of these times is the determining factor. A complete solution of this problem has been given in Ref. 17.

To simplify the analysis, it is expedient to single out two opposite cases—the cases of a narrow and of a broad spectrum of the exciting radiation.

For the present treatment we shall call a spectrum narrow when it obeys the relationship $\Gamma \gg \Delta\omega \sim 1/\tau_k$. Here the character of the process of multiphoton excitation is analogous to the direct process of multiphoton ionization of atoms that was treated above. When the above-cited inequality is satisfied, we can neglect the decay of the correlation function in Eq. (3.8), so that the probability of excitation is described by the following relationship[18]:

$$W \approx A_k G_k(0)\, \frac{2\Gamma}{(\omega_{01} - k\omega_0)^2 + \Gamma^2}. \tag{3.9}$$

Let the frequency of the monochromatic radiation equal the central frequency ω_0 of the nonmonochromatic radiation and the intensities be equal ($F_m = \langle F \rangle$). Then we can characterize the role of the nonmonochromatic radiation by the statistical factor for bound-bound transitions whenever we can neglect the frequency-dependence of the probability, just as in the case of bound-free transitions. Here we have:

$$g_k = \frac{W}{W_m} = \frac{\langle F^k \rangle}{F_m^k}.$$

If the fluctuations of the radiation are described by a Gaussian random process (as we know, this is true of the radiation from a thermal source), then the higher correlation functions can be expressed in terms of the lowest function:

$$G_k(t) = k!\,[G_1(t)]^k.$$

That is, we have

$$G_k(0) = \langle F^k \rangle = k!\,\langle F \rangle^k$$

and consequently we obtain $g_k = k!$.

Thus, for bound-bound transitions with unperturbed resonance levels and under conditions of narrow-spectrum exciting radiation, the value of the statistical factor is the same[17,18] as in the case of bound-free transitions.

We know of no direct experiments pertaining to power-function processes of multiphoton excitation of an isolated atom. However, qualitatively analogous phenomena occur for effects in atomic vapors and in excitation of molecules. As an example we can point to Ref. 20, in which sodium vapor was irradiated with dye-laser radiation and a two-photon excitation process was observed. Multifrequency radiation was employed with two different spectral widths, which were approximately 0.1 and 0.01 cm^{-1}. In the former case the width of the effective spectrum of the laser radiation was of the same order of magnitude as the Doppler absorption line of the vapor. In the latter case the spectral width of the radiation was an order of magnitude smaller than the width of the absorption spectrum, so that the case of a narrow laser-radiation

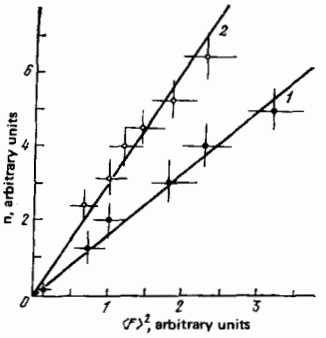

FIG. 2. Relationship of the number n of excited atoms to the radiation intensity $\langle F \rangle$ in a process of two-photon excitation with radiation having different spectral widths: 1—$\Delta\nu \approx 0.1$ cm^{-1}, 2—$\Delta\nu \approx 0.01$ cm^{-1}; the width of the absorption line ≈ 0.1 cm^{-1} (from the results of Ref. 20).

spectrum was realized. Figure 2 shows the experimental results in the form of the relationship of the number of excited atoms to the intensity of the radiation. In both cases the yield of the process is proportional to the square of the mean intensity of the radiation. However, for equal mean intensities, the yield is larger for the smaller width of exciting spectrum. The measured ratio of yields under the action of radiation with narrow and broad spectra is 1.8 ± 0.3. This agrees well with calculations by a formula such as (3.8), which give the value 1.7 ± 0.2.

Experimental data on a process of two-photon absorption by dye molecules have been obtained in Refs. 21 and 22. In both cases the statistical factor was measured, which proved close to 2! The small deviations from 2! found in Ref. 21 are hard to interpret unambiguously, since perhaps one-photon excitation processes also contribute to the yield. (The results of Ref. 22 are discussed in greater detail in Sec. 6, since the most valuable result pertains to the dependence of the statistical factor on the number of modes being generated.)

In the opposite limiting case of a broad radiation spectrum, in which the relationship $\Gamma \ll \Delta\omega$ obtains, the frequency-dependence $W(\omega)$ in Eq. (3.8) is substantially determined by the character of the decay of the correlation function $G_k(t)$. As Eq. (3.8) implies, in this case $W(\omega)$ is determined by the Fourier transform of the correlation function $G_k(t)$. That is, it is determined by the quantity $S_k(\omega) = \int_{-\infty}^{\infty} G_k(t) e^{-i\omega t} dt$. Since $F(\omega) = \int_{-\infty}^{\infty} G_1(t) e^{-i\omega t} dt$ is the spectral intensity distribution of the laser radiation, we can naturally call the quantity $S_k(\omega)$ the effective intensity spectrum corresponding to the k-photon process. Whenever the fluctuations of the radiation amount to a Gaussian random process, the kth-order effective spectrum is described by the k-fold convolution of the radiation spectrum $F(\omega)$.[23] If here the spectrum $F(\omega)$ of the radiation has a Gaussian form, then the effective kth order spectrum $S_k(\omega)$ also has a Gaussian form with a width larger by a factor of $\sqrt{k}$.

Although formally one can introduce the statistical factor for the case of a broad spectrum,[19,24] yet here the factor loses its universality, since it is determined not only by the properties of the radiation, but also by the properties of the atomic system. In particular, the ratio of the probabilities can be either larger or smaller than unity.

4. PERTURBATION OF THE ATOMIC SPECTRUM IN A NONMONOCHROMATIC FIELD

As we have stated above, apart from the high probability of multiphoton transitions, the action of a strong light field on an atom is manifested also in a substantial perturbation of the atomic spectrum that is expressed in shifts and broadenings of the atomic levels. The perturbation of the atomic spectrum has been described in rather great detail in the scientific literature in the case of a monochromatic external field.[2,3,25,26] As a rule, two limiting cases have been singled out—resonance and nonresonance perturbation. A perturbation is commonly termed a resonance perturbation whenever the matrix element governing the mixing of pairs of resonance levels exceeds the infinite sum of matrix elements that characterize the nonresonance interaction. In practice, this corresponds to having the frequency of the external field close to a transition frequency in the spectrum of the atom.

Below we shall adhere to the traditional classification of perturbations as resonance and nonresonance cases. However, in a nonmonochromatic field, this classification is considerably less sharp than in a monochromatic field, owing to the finite width of the spectrum of the radiation. Actually, as one increases the detuning between the central frequency ω_0 of the spectrum of the radiation and the central frequency of the atomic transition, a situation can arise in which the external field exerts both resonance and nonresonance types of action on the studied pair of levels.

Just as in other cases, the manifestation of nonmonochromaticity of the exciting field in the perturbation of atomic levels depends substantially on the spectral width of the radiation, i.e., on the correlation time of the laser radiation. The intensity of the nonmonochromatic field, being a random function of the time, can be considered practically invariant only over a time interval that does not exceed the correlation time τ_{cor}. During this time interval, we can speak of the energy of the atom with an accuracy no greater than $\hbar/\tau_{cor}$ (on the basis of the time-energy uncertainty relationship). If this energy uncertainty is smaller than the shift in the atomic level, i.e.,

$$\frac{\hbar}{\tau_{cor}} < \delta E_n, \tag{4.1}$$

then the shift can be fixed. At each instant of time it is determined by the instantaneous value of the intensity. That is, the shift "follows" the variation of the intensity. Yet if the energy uncertainty exceeds the shift:

$$\frac{\hbar}{\tau_{cor}} > \delta E_n, \tag{4.2}$$

then the energy of the atomic level does not follow the intensity variations, and a shift is realized that corresponds to the mean value $\langle F \rangle$ of the intensity.

In frequency language, the condition (4.1) corresponds to the case of a narrow spectrum of the perturbing field:

$$\Delta\omega < \frac{\delta E_n}{\hbar} = \delta\omega_n.$$

Here $\delta\omega_n$ is the shift of the atomic level expressed in frequency units; the condition (4.2) corresponds to a broad radiation spectrum.

We have assumed above that τ_{cor} and $1/\delta\omega_n$ are much smaller than the lifetime of the atom in the excited state with respect to the process of spontaneous relaxation. Evidently, this assumption is fulfilled in practice in all the cases of interest to us.

a) Resonance perturbation of the atomic levels

At first glance, the simplest system to examine is the two-level system: a ground state and an excited state in an external field at resonance. However, as we know,

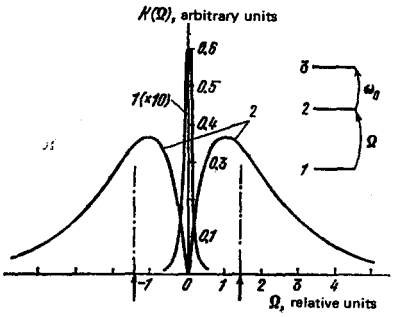

FIG. 3. Shape of the absorption line $K(\Omega)$ of a weak field at frequency Ω for an atom in the presence of a strong resonance field of frequency ω_0 (diagram of levels at upper right), according to the data of Ref. 28. 1—case of a broad spectrum of the perturbing radiation; 2—case of a narrow spectrum. The arrows indicate the positions of the components of the absorption line of the atom in the presence of the strong resonance monochromatic field at the same intensity as in case 1.

the theoretical analysis of this system requires taking into account the variation of the population of the resonance levels, which is a complicating circumstance. One must employ the language of the density matrix for a mathematical description of this case.

We can trace the fundamental regularities of the phenomenon under consideration considerably more simply on the example of a three-level system in which a double optical resonance arises.[1)] Let us treat the case in which there are two external fields— a strong field at the frequency ω_0, which is close to the frequency of the transition 2–3, and a weak "probe" field at the frequency Ω, which is close to the frequency of the transition 1–2 (Fig. 3). Here one poses the problem of describing the absorption of the probe radiation as a function of the various properties of the strong field $\mathscr{E}(t)$ and of the two-level system 2–3. The shift of the levels is manifested in the form of the absorption line of the probe field. When the problem is posed in this way, there is no need to take into account the variation of the populations of the atomic levels. This substantially simplifies the analysis.

There is at present no theoretical description of the perturbation of an atomic level in the case of an arbitrary random field $\mathscr{E}(t)$. Results have been obtained for two special cases: for a field $\mathscr{E}(t)$ that amounts to a discontinuous Markov process,[27] i.e., a process in which the amplitude and phase that characterize the nonmonochromatic radiation vary in jumps at random instants of time, and for a Gaussian field,[28] i.e., in a special case and for the radiation from a thermal source. The results for the limiting cases of broad and narrow radiation spectra proved to be qualitatively analogous. Here a radiation spectrum is broad if $(\Delta\omega)^2 \gg |V_{32}|^2$, where $V_{32} = |d_{32}|\mathscr{E}$ is the matrix element that describes the process of resonance mixing of the states 2 and 3, and it is narrow when the sense of the inequality is reversed. With a broad radiation spectrum, the absorption spectrum of the auxiliary light at the 1–2 transition is Lorentzian in shape with a width $\sim |V_{32}|^2/\Delta\omega$, which depends on the intensity and on the spectrum of the radiation (Fig. 3). With a narrow spectrum, the shape of the absorption line of the auxiliary light reflects the set of positions of the atomic level that are realized in the ensemble of random values of the field amplitude. In this case, in order to describe the shape of the line in the nonmonochromatic field, one must average the expression for the shape of the line in a monochromatic field over the amplitude distribution of the field $\mathscr{P}(A) = 2AP(A^2)$, where $A = \sqrt{F}$. Here the absorption line has the shape of a two-humped curve with a gap in the center. This is qualitatively analogous to the line splitting in resonance mixing in a monochromatic field[2] (Fig. 3). Here the spectrum of the laser radiation proves to be unessential. As we shall see from the description below, a qualitatively similar situation is always characteristic of a narrow laser-radiation spectrum. We note that the character of the random variation of $\mathscr{E}(t)$ is manifested in the transition region from a narrow to a broad spectrum.[29]

Now let us turn to the case of resonance perturbation of atomic levels in which a strong nonmonochromatic field exists in resonance with a transition of the atom from the ground state 1 to 2, while the weak field is responsible for the transition 2–3 (see the level scheme in Fig. 4). As we have stated, the change in populations of the resonance states is considerable, and saturation occurs, which must be described in the language of the density matrix. Owing to mathematical difficulties, the theoretical analysis of this case has been performed only numerically under conditions of a narrow radiation spectrum.[30] Figure 4 shows the results of calculations of the shape of the absorption line of the weak field when the strong field is at exact resonance. The strong nonmonochromatic field is described by a discontinuous Markov process with a randomly fluctuating amplitude. For comparison, the same diagram shows the results of calculations corresponding to excitation with radiation having a constant amplitude. As we should expect, the absorption line is split for a narrow spectrum of the perturbing radiation (Fig. 4) analogously to the splitting in a monochromatic field.[2] We must note that the width of the maxima is broader for a nonmonochro-

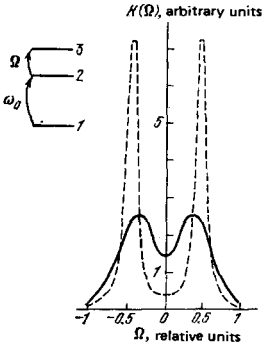

FIG. 4. Shape of the absorption line $K(\Omega)$ of a weak field at frequency Ω for an atom in the presence of a resonance field of frequency ω_0 (diagram of levels above), according to the data of Ref. 30. Solid curve—nonmonochromatic field, dotted—constant-amplitude-field.

[1)]The perturbation of the levels of the atom in this case is sometimes called the Autler–Townes effect.

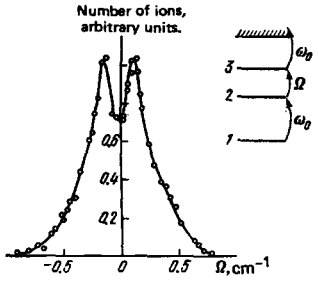

FIG. 5. Shape of the absorption line of a weak field at frequency Ω (transition 2–3 in the diagram at upper right) in the presence of a strong resonance field at frequency ω_0, according to the data of Ref. 31. The number of ions in this experiment is proportional to the number of atoms in state 3.

matic field than for a monochromatic field, while the spacing between them is smaller. The overall character of the phenomenon is analogous to the case of a double optical resonance (cf. Fig. 3). The data obtained for the nonmonochromatic field qualitatively correspond to the result of averaging over the amplitude distribution of the field.

The role of nonmonochromaticity of the laser radiation in the resonance perturbation of atomic levels has been studied experimentally on the example of the sodium atom under conditions in which the strong laser radiation field was in resonance with a transition of the atom from the ground state[31] (Fig. 5). Thus the population of the resonance levels 1 and 2 proved essential. The perturbation of the atomic level 2 was analyzed from the shape of the absorption line for the transition of the atom from 2 to 3 when acted on by the weak radiation field of a second laser that was tunable in frequency. The number of atoms that had undergone transition to the state 3 was determined by observing the ionization from this state under the action of the strong field. The width of the spectrum of the strong radiation field was small, being ≈ 0.03 cm^{-1} (about 10 longitudinal modes), whereas its field intensity was so large that the scale of the perturbation considerably exceeded the width of the spectrum. That is, the case of perturbation by narrow-spectrum radiation obtained. Figure 5 shows the dependence of the ion signal on the frequency of the weak radiation field, which reflects the resonance perturbation of level 2. We distinctly observe the splitting of the line, which corresponds to the case of a narrow spectrum of the perturbing field. The authors of Ref. 31 also obtained experimental data on the variation of the shape of the absorption line of the weak radiation field with detuning of the strong perturbing field. The results that they obtained qualitatively agree with the calculations.[30] However, a quantitative comparison is impeded by the need to take the spatial inhomogeneities of the perturbing field into account.

b) Nonresonance perturbation of the atomic levels

The character of the nonresonance perturbation of the atomic levels in a monochromatic field is rather well known.[2,25] The shift of the level n is described by a power-series expansion in the intensity of the external field:

$$\delta E_n = \alpha_{1n} F + \alpha_{2n} F^2 + \ldots.$$

Here α_{1n} is the dynamic polarizability, and α_{2n} is the dynamic hyperpolarizability of the atom. Henceforth we shall restrict the treatment to the case in which the shift is determined only by the first term of the expansion (4.3).[2)] As a rule, this is precisely the case realized in practice at a field intensity $\mathscr{E} \ll \mathscr{E}_{at}$.

One can perform a theoretical analysis of the role of nonmonochromaticity of the external field in nonresonance excitation of the atomic levels on the example of the three-level system that we studied above in describing resonance perturbation. The condition for realizing nonresonance perturbation of the levels 2 and 3 (Fig. 3) consists in having both the intrinsic width Γ_{23} of the 2–3 transition and the matrix element of the interaction between these levels much smaller than the detuning Δ of the resonance between the frequency of the external field and the frequency of the 2–3 transition: $\Gamma_{23}, |V_{32}| \ll \Delta = |\omega_{32} - \omega_0|$. When this condition is satisfied, we can derive from Eqs. (4.5)–(4.7) an expression for the shape of the absorption line of the weak probe light that manifests the shift of the levels. Just as in the earlier case, we shall single out the cases of narrow and broad spectra of the perturbing radiation.

The criterion for realizing the case of a narrow spectrum has the form

$$\frac{\alpha_{12}\langle F\rangle}{\hbar} = \delta\omega_2 \gg \Delta\omega. \tag{4.4}$$

When this criterion is satisfied, we obtain the following relationship for the shape of the spectrum:

$$K(\Omega) \sim P\left(-\frac{\omega_{21}-\Omega}{\alpha_{12}/\hbar}\right), \qquad \frac{\Omega-\omega_{21}}{\alpha_{12}/\hbar} > 0. \tag{4.5}$$

Just as in the case of resonance perturbation, the shape of the line reflects the set of positions of the atomic level that are realized in the ensemble of random values of the radiation intensity. It can be obtained by averaging the shape of the line in a monochromatic field over the distribution $P(F)$ (Fig. 6). We note that the shape of the line does not depend here on the shape of the spectrum of the laser radiation.

The criterion for realizing the case of a broad spectrum of the perturbing radiation is evidently the opposite of that for a narrow spectrum in (4.4): $\delta\omega_2 \ll \Delta\omega$. In this case the shape of the absorption line is Lorentzian with a width $\sim (\delta\omega_2)^2/\Delta\omega$ (Fig. 6). Its maximum is displaced by the magnitude of the mean shift $\delta\omega_2$.[33] We note that above we have neglected the dependence of the dynamic polarizability on the frequency of the laser radiation. In fact, such an assumption may not be valid at frequencies close to resonance, where the dynamic polarizability varies sharply with varying frequency. Here a nonmonochromaticity of the field blurs out the resonance maxima.[32]

The analysis that we have made allows us to point

[2)] The perturbation of the atomic levels in this case is sometimes termed the quadratic Stark effect in an alternating field.

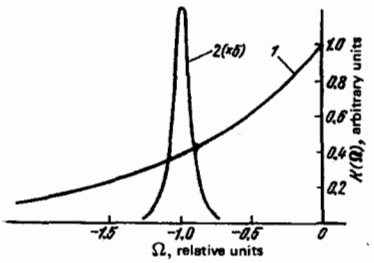

FIG. 6. Shape of the absorption line $K(\Omega)$ of a weak field at frequency Ω for an atom in the presence of a strong nonresonance field. 1—case of a narrow spectrum of the perturbing field; 2—case of a broad spectrum. The natural width of the perturbed level is assumed infinitesimally small.

out the fundamental qualitative difference of nonresonance perturbation in a nonmonochromatic field from the case of a monochromatic field—the perturbation can be described as a shift in the level only for a broad radiation spectrum. It is not expedient to employ the term "level shift" in the narrow-spectrum case, since the center of gravity of the absorption line is altered by a magnitude comparable with that of the broadening of the line. In any case, this statement is valid for the exponential distribution $P(F)$ that is realized in practice.

The experimental observation of nonresonance perturbation of atomic levels by a laser radiation field is considerably complicated by the spatial and temporal inhomogeneity of the intensity distribution of the laser radiation. The authors of Ref. 33 were able to avoid these difficulties. Both cases of broadening of atomic levels were observed experimentally upon perturbing the spectrum of the cesium atom by an external nonmonochromatic UHF field. The use as a radiation source of a UHF noise generator enabled the authors[33] to create a field that was homogeneous in the interaction volume and in time. Figure 7 shows the results of these experiments. Good quantitative agreement with the calculations is observed.

The treatment that we have carried out above of perturbation of atomic levels in a nonmonochromatic field allows the general conclusion that one can reconstruct the properties of the radiation from perturbation data

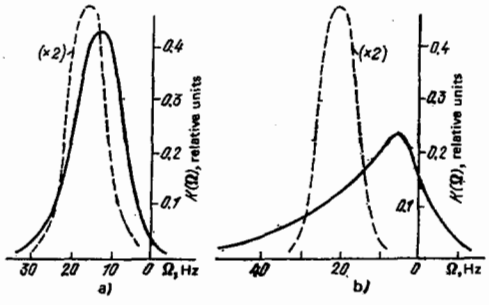

FIG. 7. Shape of the absorption line $K(\Omega)$ of a weak field at frequency Ω for an atom in the presence of a strong nonresonance field according to the data of Ref. 33. 1—case of a broad spectrum of the perturbing field ($\Delta\omega = 30$ Hz), 2—case of a narrow spectrum ($\Delta\omega = 2$ Hz). Dotted line—case of a monochromatic field.

in the narrow-spectrum case: in the resonance case one can reconstruct the field amplitude distribution $\mathcal{P}(A)$, and the intensity distribution $P(F)$ in the nonresonance case.

5. MULTIPHOTON EXCITATION OF ATOMS

Above in Sec. 3 we have discussed the power-law process of excitation of an atom in which the probability was assumed proportional to F^k, where k is the number of photons absorbed in the transition. Such a process occurs if the shifts and widths of the atomic levels induced by the field are smaller than the natural widths. In the opposite case of strong perturbation of the atomic spectrum, in which $\delta\omega(F)$, $\Gamma(F) > \Gamma$, the induced shifts $\delta\omega(F)$ and widths $\Gamma(F)$ essentially govern the process of multiphoton excitation. Generally the latter is not of a power-law type. Here the nonmonochromaticity of the exciting field is manifested both in perturbation of the resonance states and in the probabilities of the multiphoton transitions. Just as was done above, it is also expedient in treating multiphoton excitation in a nonmonochromatic field to single out the cases of narrow and broad spectra of the exciting radiation.

The condition for realizing a narrow spectrum has the form

$$\Delta\omega \ll \Gamma(F),\ \delta\omega(F). \tag{5.1}$$

The decisive factor is the greater of the two quantities standing on the right-hand side of the inequality (5.1). As we have noted above, in the case of a narrow spectrum, one must use the value of the probability in a monochromatic field averaged over the intensity distribution $P(F)$ to describe the probability of the nonlinear process in a nonmonochromatic field[34,35]:

$$W \sim \int_0^\infty P(F)\, F^k \frac{\Gamma(F)}{[\omega_f - k\omega_0 + \delta\omega(F)]^2 + [\Gamma(F)]^2}\, dF. \tag{5.2}$$

Here ω_f is the frequency of the transition in the unperturbed atom.

Let us examine the special case in which the shift dominates, i.e., $\delta\omega(F) \gg \Gamma(F)$. Here Eq. (5.2) gives rise to an expression for the excitation probability that depends on the detuning of the k-photon resonance $(\omega_f - k\omega_0)$:

$$W \sim (\omega_f - k\omega_0)^k P\left(\frac{\omega_f - k\omega_0}{\alpha_{1f}/\hbar}\right). \tag{5.3}$$

Equation (5.3), which was derived in Ref. 24, is valid when two additional conditions are satisfied. First, resonance with the shifted level must be realized. That is, the relationship $(\omega_f - k\omega_0)/(\alpha_{1f}\hbar) > 0$ must hold. Moreover, the condition $|\omega_f - k\omega_0| \gg \Gamma(F)$ must be satisfied to allow one to write the value of the probability W in the analytic form of (5.3). Thus the relationship amounts to an asymmetrically broadened line (Fig. 8). In principle one can reconstruct the distribution $P(F)$ from this relationship. If one has an exponential intensity distribution and a one-photon transition from the excited state to the continuous spectrum (i.e., when $\Gamma(F) \sim F$), one can derive an analytic expression for W for an arbitrary relationship between the shift and

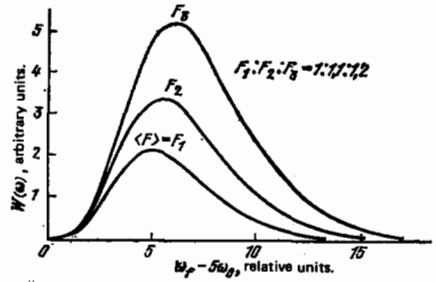

FIG. 8. Frequency-dependence of the probability of five-photon excitation of an atom in the presence of perturbation of the atomic levels (shift dominating) under conditions of narrow-spectrum laser radiation (according to the data of Ref. 34).

the broadening of the resonance state.[35] Qualitatively, the character of the $W(\omega)$ relationship in the special case that we have treated naturally matches the character of the relationship in the general case.

In the case of a broad spectrum, in which $\Delta\omega \gg \delta\omega(F)$, $\Gamma(F)$, as the qualitative analysis performed above in Sec. 4 implies, the mean shifts and mean widths of the levels must be manifested in the transition probability. There are currently no studies that have quantiatively analyzed multiphoton excitation of atoms with account taken of strong perturbation of the spectrum by a nonmonochromatic field with a broad radiation spectrum. Qualitatively there is no doubt that the width of the resonance is determined by the width of the effective spectrum of the laser radiation for k-photon excitation, while the degree of nonlinearity is determined by the quantity k, i.e., by the number of absorbed quanta. We should note that there have been no direct experiments on the role of nonmonochromaticity in multiphoton excitation under conditions of perturbation of the atomic spectrum.

6. NONLINEAR IONIZATION OF ATOMS

The character of the process of nonlinear ionization of an atom in an alternating field in which the quantum energy $\hbar\omega$ is smaller than the binding energy E_i of an electron is governed by three parameters—the intensity and the frequency of the field and the binding energy of the electron. Depending on the relationship between these quantities, the ionization process has the character of a direct multiphoton process, a tunneling effect, or a transition via an intermediate resonance state (resonance ionization process).[17] All three cases have been described in detail in many studies, while the direct and resonance processes have been studied experimentally in detail.[1-3] However, as in a number of other cases, the field was assumed monochromatic in the overwhelming majority of calculations, whereas a nonmonochromatic laser radiation field has been employed in the overwhelming majority of experimental studies. We shall assume in this section that the fundamental regularities of the process of nonlinear ionization in a monochromatic field are known,[2,3] and we shall treat the specific phenomena that arise in nonlinear ionization in a nonmonochromatic field.

a) The direct process of multiphoton ionization of atoms

As we have shown in Sec. 3, in the direct process of multiphoton ionization, taking account of the role of nonmonochromaticity is reduced to introducing the statistical factor, whose magnitude is determined by Eq. (3.4).

One takes the mean intensity $\langle F\rangle$ of the nonmonochromatic radiation in Eq. (3.4) to be the energy of the radiation per fixed interval of time that exceeds the correlation time. As a rule, the correlation time for the multifrequency laser radiation under study is shorter than 10^{-9} s.

In order to measure correctly the mean radiation intensity of a multifrequency laser, one must employ single-photon detectors that satisfy the evident requirement: the time constant of the detector must exceed by a large factor the characteristic duration of the fluctuations of the radiation. This requirement is satisfied by the standard detectors, e.g., photodiodes. In a pulsed generation regime, one must take into account also the shape of the envelope—the time constant of the single-photon detector must be shorter than the duration of the pulse. When the pulses of a single-frequency and a multifrequency laser have the same shape, the requirement of equal mean radiation intensities is equivalent to the requirement of equal energies of the radiation per pulse. If the shapes of the pulses differ, then in consideration of the nonlinear interaction that they cause, we require not that the energies be equal, but the quantities

$$\int_0^{\tau} F_{av}^k(t)\,dt = \int_0^{\tau_0} F_m^k(t)\,dt.$$

Here $F_{av}(t)$ is the envelope of a pulse of the multifrequency laser of duration τ, while $F_m(t)$ is the envelope of a pulse of the single-frequency laser of duration τ_0.

Let us turn to the experimental and theoretical data on the effect of the number and mutual coupling of the modes on the magnitude of the statistical factor.

If we start with the distribution of (2.3), then the expression for the relationship of the statistical factor to the number of modes has the form[10,11]

$$g_k = k!\,\frac{N^k(N-1)!}{(N+k-1)!}. \qquad (6.1)$$

We see from this relationship that the statistical factor increases with increasing number of generated modes, while approaching the value $k!$ asymptotically (see also Ref. 36). In the other limiting case with $N=1$, Eq. (6.1) gives the value $g_k(1)=1$ for the statistical factor. This corresponds to the value of the statistical factor for the radiation from a single-frequency laser. Figure 9 shows the relationship of the statistical factor to the number of modes for different values of the degree of nonlinearity k.

Let us formulate the criterion of resemblance of the laser radiation to that of a thermal source as a function of the number of modes. We base this on the asymptotic character of the $g_k(N)$ relationship for large N. Resemblance exists when the number of modes $N > N^*$, such that the relative deviation $g_k(N)$ from $k!$ does not

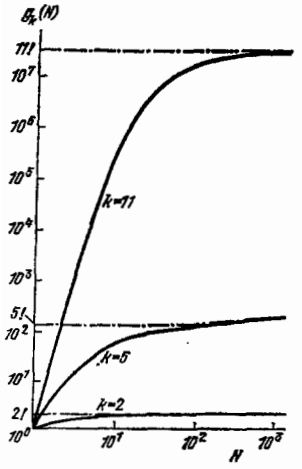

FIG. 9. Dependence of the statistical factor $g_k(N)$ on the number N of modes generated by the laser for values of the degree of nonlinearity $k=2$, 5, and 11 [calculated by Eq. (6.1)].

exceed the quantity C. That is, in line with the asymptotic behavior of (6.1), the following relationship is satisfied

$$N > N^* = \frac{k(k-1)}{2C}. \tag{6.2}$$

In practice one takes the quantity C to be the accuracy of measurement of the statistical factor g_k.

As Eq. (6.2) implies, the higher is the degree of nonlinearity k of the process, the greater is the number of modes that the multifrequency laser must generate to make the action of its radiation equivalent to that of a thermal source. When $N > N^*$ [see (6.2)], the result of the interaction depends neither on the number of modes nor on the shape of the spectrum. As is implied by all that we have said above, the criterion for realizing such a generation regime is the observation of a statistical factor $g_k = k!$.

The relationship (6.2) allows certain conclusions to be drawn concerning the expedience of employing the radiation of various multifrequency lasers for studying nonlinear effects on the basis of the width of the laser radiation spectrum and the attainable accuracy C of measuring the statistical factor. Thus, for example, the radiation spectrum of the widespread ruby laser has the width $\Delta\omega \lesssim 1$ cm^{-1}. That is, the laser emits no more than 10^2 modes. On the basis of the data of Eq. (6.2), we can treat the radiation of such a laser as equivalent to that of a thermal source only for two- or three-photon processes. A neodymium-glass laser offers considerably greater possiblities ($\Delta\omega \approx 10$ cm^{-1}, $N \sim 10^3$ modes). We can treat the radiation of this laser as being equivalent to that of a thermal source in experiments with $k \leq 20$, i.e., up to the limiting value that has been currently realized in experiments with radiation in the visible frequency range.

At present, experimental data on the $g_k(N)$ relationship have been obtained only for the radiation of the neodymium-glass laser[22,37] operating in a Q-switched regime. The results of these experiments clearly indicate that the statistical factor falls below $k!$ when there is a small number of modes such that $N < N^*$.

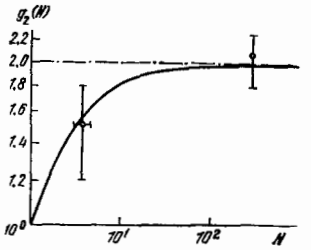

FIG. 10. Experimental values of the statistical factors $g_2(N)$ for neodymium-laser radiation for various numbers of generated modes.[22] Solid curve—calculation by Eq. (6.1).

The process of two-photon ionization was employed in Ref. 22 as the laser-radiation detector instead of the direct ionization process. The experimental data on the magnitude of the absorption obtained for $N=1$, 3–5, and 300 yield a $g_2(N)$ relationship that fits Eq. (6.1) within the limits of accuracy of the measurements (Fig. 10). However, the accuracy of this experiment is not high enough to draw any conclusions on the role of the shape of the spectrum. In Ref. 37, the process of eleven-photon ionization of the xenon atom was employed as the detector. The results of the measurements qualitatively coincide with the relationship (6.1), but there is no quantitative agreement (Fig. 11). The discrepancy can arise from various factor (cf. Ref. 38). One of them is the non-power-function character of the interaction that is characteristic of this process.[39] Moreover, nonlinear effects can also arise in the laser resonator that distort the distribution $P(F)$. However, the assumption that they play a dominant role that was expressed in Ref. 40 is not substantiated by independent experimental data.

The results of these experiments indicate the need of carrying out extensive studies of the $g_k(N)$ relationship and the promise offered by multiphoton detectors for such studies. We note that one must test independently the power-function character of the interaction of the radiation with the detector and of the spectrum of the laser radiation to allow an unambiguous interpretation of the results of such experiments.

The possibility of realizing a generation regime of

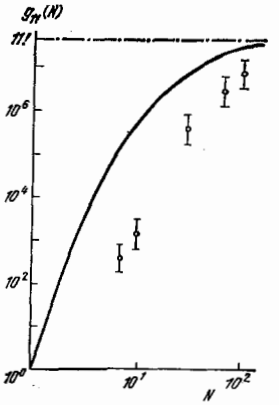

FIG. 11. Experimental values of the statistical factors $g_{11}(N)$ for neodymium-laser radiation for various numbers of generated modes.[37] Solid curve—calculation by Eq. (6.1).

many independent modes has been tested experimentally for a neodymium-glass laser for processes of degree of nonlinearity $k \leq 5$. Rather precise results have been obtained[41,22] for a pulsed generation regime with Q-switching of the resonator. In Ref. 41, data were obtained on the value of g_5 by employing a five-photon process of ionization of the sodium atom as the detector. This experiment was performed with a number of generated modes $N = 4 \times 10^3$, which is an asymptotically large value at the degree of nonlinearity $k = 5$. The value $g_5 = 10^{2.04 \pm 0.25} = 5!$ was obtained. Another experiment[22] has observed the process of two-photon absorption in the dye rhodamine 6G. The laser generated $N = 300$ modes, which is also an asymptotically large quantity for $k = 2$. The measured value was $g_2 = 2.1 \pm 0.2 = 2!$. The results of these experiments show that one can realize a generation regime of multifrequency radiation with a neodymium laser in which this radiation is equivalent with sufficient accuracy to the radiation from a thermal source for processes of degrees of nonlinearity $k \leq 5$. We should note that these data do not permit analogous conclusions to be drawn for processes having $k > 5$. Actually, interaction between the modes must lead to a larger effect with increasing k. Therefore, in principle, it may not be manifested within the experimental limits of accuracy for small k while being manifested for large k.

Observation of statistical factors equal to $k!$ is a necessary condition for equivalence of the properties of the radiation of multifrequency lasers to those of the radiation from a thermal source. Correspondingly, the requirement of a sufficiently large number of modes in (6.2) is also a necessary condition. Although these conditions are not sufficient, their fulfillment can serve in a number of cases as an indirect justification for employing the exponential intensity distribution function $P(F)$ inherent in the radiation from a thermal source in calculations describing the interaction of multifrequency laser radiation with an atom.

We must bear in mind that the measured value fluctuates in any procedure for measuring the statistical factor. For a fixed mean radiation intensity, the yield of the process fluctuates more for a smaller number of generated modes (smaller number of fluctuational excursions per axial period) owing to fluctuations of the instantaneous values. We note that the fluctuations of the yield substantially exceed those of the radiation intensity, owing to the nonlinear character of the interaction. References 11 and 13 are devoted to analyzing the dependence of the fluctuations on the degree of nonlinearity of the process and on the number of generated modes of the laser radiation. Measurements of the fluctuations of the yield of a multiphoton process of ionization of atoms have not been performed with an accuracy sufficient for comparison with the results of these calculations. The only known data show that the amplitude of the fluctuations of the yield in multimode laser radiation considerably exceeds the amplitude with single-frequency radiation.[39]

We note that, in addition to the effect on the probability of the direct ionization process, the nonmonochromaticity of the field can also alter the frequency-dependence of the ionization probability in the frequency regions in which the multiphoton cross-section has sharp inter-resonance minima.[42]

b) The tunneling effect in an alternating field

It is well known that the process of ionization of an atom in an alternating field has the character of a tunneling effect if the adiabaticity parameter is smaller than unity: $\gamma = (\omega/\mathscr{E})\sqrt{2E_n} \ll 1$. As before, $\mathscr{E}$ and ω are the intensity and frequency of the field, while E_n is the binding energy of the electron occupying the level n. In this case the ionization probability depends exponentially on the intensity of the field but does not depend on its frequency. Accordingly, the difference between ionization in nonmonochromatic and monochromatic fields at equal mean intensities reduces only to a change in the ionization probability, and does not depend on the width of the laser radiation spectrum. Thus the role of nonmonochromaticity of the radiation in tunneling ionization can be taken into account by introducing the corresponding statistical factor.

A transition from the multiphoton limiting case ($\gamma \gg 1$) to the tunneling case ($\gamma \ll 1$) at a fixed radiation frequency takes place as the field intensity increases. It is of interest to elucidate how the magnitude of the statistical factor varies here. Qualitatively it is evident that since the ionization probability increases more slowly with increasing intensity of the radiation when γ decreases in the region where $\gamma \lesssim 1$, than under conditions when $\gamma \gg 1$, then this should lead to a statistical factor depressed below the value of g_k. Actually, as we have seen above, in the case being studied, the difference between multifrequency and single-frequency radiation at a fixed mean intensity of the radiation reduces to the existence in the former case of instantaneous values of the intensity that considerably exceed the mean value. Owing to the weaker $W(F)$ relationship for $\gamma \ll 1$ as compared with the corresponding relationship for $\gamma \gg 1$, the role of large intensity excursions in the former case is relatively smaller than in the latter case. Correspondingly the statistical factor is smaller.

In order to determine the statistical factor in tunneling ionization and its dependence on the field intensity, one must derive an expression describing the tunneling probability in the nonmonochromatic field. Evidently, in this case the probability W is described by Eq. (3.2):

$$W(\langle F \rangle) = \int_0^\infty W_m(F)\, P(F)\, dF.$$

As before, $W_m(F)$ is the tunneling probability in a monochromatic field.

Upon assuming that the intensity distribution $P(F)$ of the radiation is described by the exponential function (2.2), we can calculate this integral analytically. The probability of tunneling ionization in the nonmonochromatic field is described (in atomic units) with exponential accuracy by the following expression[43]:

$$W = \exp\left(-2\sqrt{3}\,\frac{E_n}{\langle F \rangle^{1/2}}\right). \qquad (6.3)$$

We note that this relationship differs from the well-known relationship describing the probability of tunneling in an alternative monochromatic field[17]:

$$W_m = \exp\left(-\frac{4}{3}\sqrt{2}\,\frac{E_n^{3/2}}{F^{1/2}}\right). \qquad (6.4)$$

The variation given by Eq. (6.3) is smoother than W_m. As we have said above, this seems qualitatively sufficiently evident—large excursions of the radiation intensity play the major role in the nonmonochromatic field.

Although the analytic form of the relationships $W(\langle F\rangle)$ and $W_m(F)$ differs, yet as happened before for power-function relationships, in this case the difference between a nonmonochromatic and a monochromatic field can be expressed in terms of the statistical factor of (3.4):

$$g = \frac{W(\langle F\rangle)}{W_m(F)}.$$

A number of studies[39,44-47] has been devoted to calculating this factor, including the experimentally most interesting intermediate range in which $\gamma \sim 1$. Figure 12 shows the value of the statistical factor for $\gamma \sim 1$ as obtained by numerical integration for two special cases. These cases reduce for $\gamma \gg 1$ to power-function processes with the values $k=5$ and $k=11$. We see that q begins to deviate appreciably from $k!$ already at values $\gamma \gtrsim 1$. Here the deviation increases for larger k.

An experimental observation of the deviation of the statistical factor g from $k!$ has been performed[38] on the example of ionization of the xenon atom in the radiation field of a multifrequency neodymium-glass laser. For $\gamma \gg 1$ this is an eleven-photon ionization process that has been well studied experimentally. The design of the experiment to measure g was analogous to that for measuring the statistical factor g_5 (see Sec. 6.a). Here the same two lasers were used, operating in the same regimes—single-frequency and multifrequency. In the latter case the laser emitted 4×10^3 longitudinal modes. In line with Eq. (6.2), this value was high enough that the value of the statistical factor $g_{11}=11!$ could be realized with good accuracy for the eleven-photon ionization process. Ionization was observed at a field intensity $\mathscr{E} \approx 5\times10^7 \mathrm{V\cdot cm^{-1}}$, which corresponds to the value $\gamma \approx 5$. Control experiments in which the frequency of the laser radiation was varied showed that intermediate resonances play no role. Figure 12 shows the experimental results. We see that the value $g = 10^{5\pm1}$ is two orders of magnitude smaller than $g_{11} = 10^{7.6}$. The experimentally measured value of g agrees well with the calculated data.

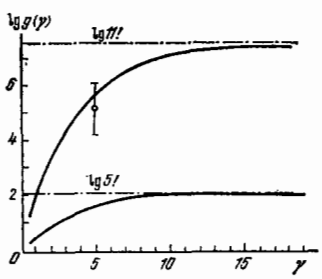

FIG. 12. Dependence of the statistical factor on the parameter γ in two special cases of ionization of atoms that reduce for $\gamma \gg 1$ to power-function processes with $k=5$ and $k=11$ Solid curves—calculation[46]; circle—experimental value.[3]

As we have noted above (see Sec. 6, a), the experimental data[37] yielded a different value of the statistical factor than is predicted by Eq. (6.1). Here observations were made of the same process of ionization of the xenon atom by light from a neodymium-glass laser, which emitted a different small number of longitudinal modes. One of the reasons might be the small value of the parameter γ, which was the same in the experiments of Ref. 37 as in Ref. 39, i.e., $\gamma \sim 5$.

In closing we note that, if we remain within the framework of the optical frequency range and of neutral atoms existing in the ground state, the condition $\gamma \ll 1$ can be realized only at a field intensity $\mathscr{E} \gtrsim \mathscr{E}_{at}$. However, the general theory of ionization[17] is valid only for $\mathscr{E} \ll \mathscr{E}_{at}$. This reflects the well-known fact that the atom practically instantaneously (within the atomic time of $\sim 10^{-17}$ s) ceases to exist as a bound system at $\mathscr{E} \sim \mathscr{E}_{at}$. Therefore the deviation of g from g_k is of fundamental interest as the limit below which the factorial expression for the statistical factor is valid.

We must also bear in mind that a value of the parameter $\gamma \ll 1$ can also be realized when $\mathscr{E} \ll \mathscr{E}_{at}$ in the case of ionization from highly excited states (small E_n) in a field in the infrared frequency range (small ω). Thus the value of the statistical factor can differ considerably from $k!$.

c) The resonance process of ionization of an atom

The process of multiphoton ionization of an atom is commonly termed a resonance process whenever the energy of a transition allowed by the corresponding selection rules from the initial state E_0 to any excited, bound electronic state E_1 matches the energy of one or of several quanta of the external field (Fig. 13). The criterion for realizing resonance with an isolated level has the form

$$|\Delta|, \Gamma \ll |E_1 - E_0|.$$

The detuning of the resonance

$$\Delta = E_1 - E_0 - k_1\hbar\omega$$

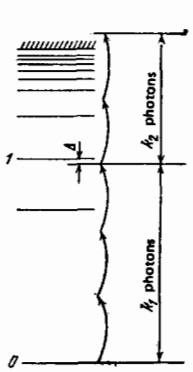

FIG. 13. Diagram of atomic levels explaining the conditions of the resonance ionization process, $k = k_1 + k_2$.

and the width Γ of the resonance state must be small compared with the energy of the resonance transition. The strongest manifestation of resonance is usually restricted to a range of detuning that does not exceed the width of the resonance level:

$$|\Delta| \leqslant \Gamma.$$

Here, whenever we are dealing with strong fields, both the detuning of the resonance and the width of the resonance state are functions of the field intensity.

In a weak monochromatic field in which the shift and broadening of the resonance state are smaller than its natural width, the probability of resonance ionization is described by the well-known formula[2]:

$$W_m = \frac{2\pi}{\hbar} \frac{|V_{01}^{(k_1)}|^2}{\Delta^2+\Gamma^2} |V_{1E}^{(k_2)}|^2 \sim F^k. \tag{6.5}$$

Here the V are the matrix elements of the transitions, and Γ is the natural (spontaneous) width of the resonance state.

The relationship (6.5) is not valid for strong fields, in which the shift and/or the broadening of the resonance state, being *per se* functions of the field intensity, are larger than its natural width. It is well known that three different physical phenomena can perturb the resonance state. They are, first, the mixing of the ground and resonance states, which leads to appearance of the so-called field width of the resonance:

$$\Gamma_f = d_{01}\mathscr{E}^{k_1} = d_{01}F^{k_1/2}. \tag{6.6}$$

Second, there is the nonresonance shift of the states 0,1, which alters the transition energy by the amount

$$\delta\omega_{01} = \frac{1}{4}\alpha\mathscr{E}^2 = \frac{1}{4}\alpha F. \tag{6.7}$$

Finally and third, there is the broadening of the resonance state owing to transitions of an electron to the continuous spectrum—the so-called ionization broadening:

$$\Gamma_i = \alpha_{1E}\mathscr{E}^{2k_2} = \alpha_{1E}F^{k_2}. \tag{6.8}$$

In (6.6)–(6.8), d_{01} is the matrix element of the dipole moment for $k_1=1$ and the corresponding compound matrix element for $k_1>1$; α is the difference between the dynamic polarizabilities of the ground and resonance states; α_{1E} is the cross-section for the process of ionization from the resonance state. In the relationship for the nonresonance shift $\delta\omega_{01}$, we have given only the first term, quadratic in the field intensity, from the expansion of the dynamic polarizability. As a rule, the restriction to the first term of the expansion is valid at not very high field intensities $\mathscr{E} \ll \mathscr{E}_{at}$, with the exception of narrow spectral intervals in the inter-resonance intervals where the dynamic polarizability changes sign in pass through zero.[2,3] We note that the functional dependence of the field perturbation Γ_f and the ionization perturbation Γ_i is determined in each concrete situation by the multiphoton character of the corresponding transitions k_1 and k_2. Depending on the values of k_1 and k_2, some particular perturbation process may dominate, or else one may not be able to single out the dominant process on the basis of the analytic relationships; sometimes one can do this via numerical estimates of the corresponding matrix elements. Many studies have been devoted to describing the process of resonance ionization in a strong monochromatic field, and their results are summarized in Refs. 2 and 48. In most cases, the probability of ionization in a strong monochromatic field in the presence of an intermediate resonance is described by a relationship of the type of (6.5). Here the role of Γ is played by a combination of the quantities Γ_f and Γ_i, while the quantity $\delta\omega_{01}$ is added to the detuning Δ. Certain special cases exist in which the relationship for the probability of resonance ionization has a different, more complicated character.[48] However, we shall not treat them, since their probability of realization in practice is small.

The effect of the perturbation of the resonance state is reduced to: a) a shift of the resonance maximum as a function of the probability of the frequency of the field $W(\omega)|_{\langle F\rangle = \text{const}}$; b) a change in the width of the resonance compared with the weak-field case; c) observation of a $W(\langle F\rangle)|_{\omega = \text{const}}$ relationship either more or less steep than the original power-function relationship for the weak-field case where $W \sim \langle F\rangle^k$; d) inapplicability of a simple formula of the type of (6.5) in a number of cases.

In resonance ionization in a weak field in which the perturbations of the resonance state are smaller than its natural width, nonmonochromaticity of the external field affects only the character of transitions of the electron from one state to another. In resonance ionization in a strong field, nonmonochromaticity of the external field is also manifested in the character of the perturbation of the resonance state. In contrast to resonance ionization in a monochromatic field, the new effects inherent in a nonmonochromatic field involve both the random nature of the intensity distribution of the external field in time and the finite width of the radiation spectrum. We shall not consider the finite duration of the radiation pulse, whose manifestation was taken into account in describing the resonance ionization process,[48] since the reciprocal of the pulse duration for multifrequency lasers is smaller than the width of the spectrum.

We shall treat the process of resonance ionization in a nonmonochromatic field in two limiting cases— in which the width of the radiation spectrum is greater or smaller than the perturbation of the resonance state.[49] Just as in the case of a monochromatic field, the governing factor is the larger of the widths that occurs.

We shall term a radiation spectrum broad if it satisfies the relationship:

$$\Delta\omega \gg \max\{\Gamma_f, \Gamma_i, \delta\omega_{01}, \Gamma\}. \tag{6.9}$$

A narrow spectrum corresponds to the converse relationship being satisfied:

$$\Delta\omega \ll \max\{\Gamma_f, \Gamma_i, \delta\omega_{01}\}. \tag{6.10}$$

We note that the natural width Γ of the atomic level has been omitted in the latter inequality. This is because, if we restrict the treatment to the interaction of nonmonochromatic laser radiation with isolated atoms and molecules, then the condition $\Delta\omega \ll \Gamma$ is not

realized in practice. The realization of the inequality (6.9) or (6.10) is governed both by the width of the spectrum and by the radiation intensity, since the amplitude of the perturbation of the resonance state depends on the radiation intensity. For a fixed width of the radiation spectrum, increasing the field intensity broadens the resonance state, so that the broad-spectrum case of (6.9) can reduce to the narrow-spectrum case of (6.10).

A broad radiation spectrum is characterized by fluctuations of the radiation intensity that are fast ($\tau_{cor} \approx 1/\Delta\omega$) in comparison with the times $1/\Gamma_f$, $1/\Gamma_i$, and $1/\delta\omega_{01}$. Hence the perturbation of the resonance state is determined by the averaged characteristics of the external field (see Sec. 4). Correspondingly, the following relationships hold:

$$\Gamma_f = d_{01}\langle F^{k_1/2}\rangle, \tag{6.11}$$

$$\delta\omega_{01} = \frac{1}{4}\alpha\langle F\rangle, \tag{6.12}$$

$$\Gamma_i = \alpha_{1E}\langle F^{k_2}\rangle. \tag{6.13}$$

A narrow radiation spectrum is characterized by fluctuations of the radiation intensity that are slow in comparison with the times $1/\Gamma_f$, $1/\Gamma_i$, and $1/\delta\omega_{01}$. Hence the perturbation of the resonance state is described by relationships analogous to those for a monochromatic field, (6.6)–(6.8), in which the instantaneous values of the radiation intensity $F(t)$ are involved.

Qualitatively, the case of a narrow spectrum of nonmonochromatic radiation is analogous to the case of a monochromatic field with a slowly varying amplitude of the intensity (compared with the characteristic time scale of the perturbation process). Correspondingly, in the narrow-spectrum case, just as in the case of monochromatic radiation, the presence of a perturbation of the resonance state determines the character of the dependence of the ionization probability on the intensity and frequency of the radiation.

In order to determine the probability W of ionization in a nonmonochromatic field under the action of narrow-spectrum radiation, we must average the value of the probability in a monochromatic field over the radiation intensity distribution $P(F)$ of the nonmonochromatic field:

$$W = W(\langle F\rangle, \omega) = \int_0^\infty W_m(F, \omega)\, P(F)\, dF. \tag{6.14}$$

In (6.13), $W_m(F, \omega)$ is the ionization probability in a monochromatic field. In a narrow-spectrum nonmonochromatic field, the role of the frequency ω is played by the central frequency ω_0 of the radiation spectrum, while $\langle F\rangle$ is a parameter of the distribution $P(F)$. The fact that the radiation spectrum in Eq. (6.14) is characterized only by the central frequency ω_0 reflects the specifics of the narrow-spectrum case, in which the shape of the radiation spectrum plays no role.

The process of resonance ionization in a broad-spectrum nonmonochromatic field differs in that the ionization arises under the action of the radiation with unchanged spectral characteristics, in spite of the change in the central frequency and shape of the spectrum of the resonance transition. This is because the perturbation is far smaller than the width of the spectrum. Thus the perturbation of the resonance state does not alter the relationships $W(\langle F\rangle)|_{\omega=\text{const}}$ and $W(\omega)|_{\langle F\rangle=\text{const}}$. The dependence of the ionization probability on the radiation intensity is described by the power-function relationship $W \sim \langle F^k\rangle$, while the frequency-dependence of the probability is described by the effective kth-order radiation spectrum. We note that the upper bound of the interval of variation of the radiation intensity in which the power-function relationship $W \sim \langle F^k\rangle$ holds is determined, on the basis of the inequality (6.9), by the width of the radiation spectrum. The larger is $\Delta\omega$, the higher is this bound.

The process of resonance ionization can be employed to study the character of a perturbation of the resonance levels. Here the ionization from the excited state serves only as a method for detecting excited atoms (the pertinent experiments are discussed in Ref. 25). Most of such experiments have been performed with nonmonochromatic laser radiation. For a number of reasons, their quantitative interpretation is difficult. First, a number of the experiments pertain to the intermediate case of interaction (between the narrow and broad spectra), whose description involves considerable difficulties. Second, quantitative comparison of the experimental results with the theory is complicated by the need to take into account the spatial inhomogeneity and shape of the pulse envelope of the laser radiation.

1) *Resonance ionization by narrow-spectrum radiation.* The probability of resonance ionization with a narrow radiation spectrum in (6.14) is determined by the probability of ionization in a monochromatic field averaged over the intensity distribution $P(F)$. Upon assuming that the distribution $P(F)$ is described by the exponential function of (2.2), a number of studies have carried out the averaging procedure on various concrete cases of perturbation of the resonance state with various degrees of nonlinearity of the transitions.[34,50–53] We note that one must always bear in mind the space-time inhomogeneity of the distribution of the laser radiation in the region of formation of the ions when comparing the results of such an averaging with the experimental data. The inhomogeneity substantially smooths out the distribution $W(\omega)|_{\langle F\rangle=\text{const}}$, and alters the $W(\langle F\rangle)|_{\omega=\text{const}}$ relationship and the absolute value of the ionization probability.

As is well known, the nature of the $W_m(F)$ relationship in a monochromatic field in the presence of a perturbation of the resonance state larger than its natural width depends on the magnitudes of k_1 and k_2, and can vary in character.[48] For certain values of k_1 and k_2, one can single out some particular dominant process of perturbation of the resonance state. Thus, when $k_1 = 1$ with an arbitrary value of k_2, the dominant process is the resonance mixing of the states 0, 1, which gives rise to a field width of the resonance state of $\Gamma_f \sim \sqrt{F}$. When $k_1 > 2$ and $k_2 > 1$, the dominant process is the nonresonance mixing of the levels 0, 1 (if we restrict ourselves to taking into account only the first term of the expansion of the dynamic polarizability in terms of the field

intensity), and it alters the energy of the 0,1 transition by the amount $\delta E \sim F$. In these two cases one can unambiguously analyze the role of the perturbation. One can conduct such an analysis for other values of k_1 and k_2 only by taking into account the quantitative relationships between the matrix elements describing the various perturbing processes, since their field-dependences are the same. Sometimes such an analysis also yields valuable results, e.g., in the practically important case in which the ionization process from the excited state is of a single-photon type ($k_2 = 1$) while excitation is substantially multiphoton ($k_1 > 2$).

Let us examine the cases in which one can single out a certain dominant perturbation process.

a) $k_1 = 1, k_2$ *arbitrary*, single-photon resonance between the states 0,1; the perturbation of the resonance state is determined by the resonance mixing of these states. The special case in which $k_2 = 1$ (two-photon ionization process with a one-photon resonance) has been treated in Refs. 50, 53, and 54. The existence of resonance mixing (saturation of the 0,1 transition) has the result that the populations of the states 0 and 1 are approximately equal, and the resonance ionization process is governed only by the transition to the continuous spectrum.[48]

In this case one must substitute into Eq. (6.14) the well-known expression for the probability $W_m(F, \omega)$ in a monochromatic field. Such a procedure for the radiation from a thermal source has been carried out[50] for $k_2 = 1$. The result of this procedure is not expressed in elementary functions, and is given in Fig. 14. The $W(\omega)$ relationship amounts to a resonance maximum of width $\Gamma_f \approx d_{01}\sqrt{\langle F \rangle}$. The fluctuations of the amplitude of the nonmonochromatic field cause the width of the resonance to be somewhat greater than in a monochromatic field. In the special case of $k_2 = 1$ at exact resonance, the statistical properties of the radiation in two-photon resonance ionization are not manifested.

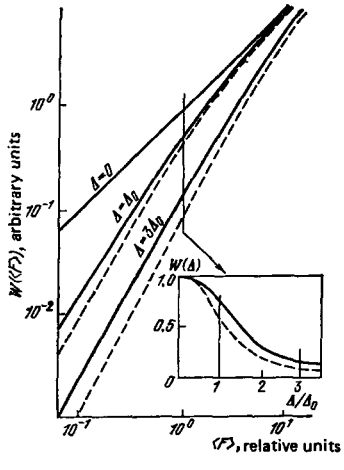

FIG. 14. Dependence of the probability of two-photon ionization of an atom ($k_1 = 1$, $k = 2$) on the mean intensity of nonmonochromatic radiation (solid curves) for various detunings (according to the data of Ref. 50). Dotted curves—the same for monochromatic radiation. Inset—frequency-dependence of the ionization probability.

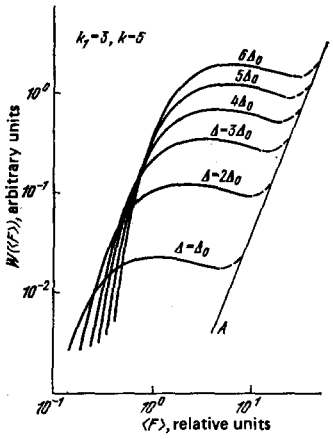

FIG. 15. Dependence of the probability of five-photon ionization of an atom having a three-photon resonance ($k_1 = 3$, $k = 5$) on the mean intensity of nonmonochromatic radiation for various detunings Δ. Line A—contribution of the direct process to the ionization probability.

b) $k_1 > 2, k_2 > 1$; the perturbation of the resonance state is governed by the process of nonresonance shift of the ground and excited states. The dependence of the probability on the intensity is determined by the initial detuning of the resonance. If the detuning has a value such that the mean shift of the levels leads to tuning the resonance, then the $W(\langle F \rangle)$ relationship becomes faster than $W \sim \langle F \rangle^k$. If the mean shift detunes the resonance, then $W(\langle F \rangle)$ becomes more gradual (Fig. 15). We should bear in mind the fact that the realization of different amplitudes of shifts under conditions of a narrow radiation spectrum corresponds to the different instantaneous values of the radiation intensity. Thus a broadening of the resonance state arises that is inhomogeneous in time. At a value of the mean shift $\delta\omega_{01} > \Delta\omega$, the magnitude of the broadening evidently weakens the effect of the detuning of the resonance on the $W(\langle F \rangle)$ relationship.

The dependence of the ionization probability on the frequency $W(\omega)$ is of a rather complicated nature. On the one hand, the maximal shifts $\delta E \sim F$ (where F is the instantaneous value of the radiation intensity) that are realized in the maximal field correspond to the maximal probability of the two transitions and hence also to the maximal ionization probability. On the other hand, in the radiation field of a multifrequency laser, the probability of fluctuational excursions of maximal amplitude is exponentially small. Moreover, the initial detuning of the resonance is of substantial importance, since it determines the fields that contribute most to the ion yield—the greater is the detuning, the higher is the intensity of the "effective" field. The joint action of these factors gives rise to the $W(\omega)$ relationship. Thus, on the frequency scale, the resonance distribution $W(\omega)$ lies on one side of the resonance frequency for zero field. The distribution broadens with increasing mean intensity of the radiation, while its shape is determined by the shape of the distribution $P(F)$ and the degree of nonlinearity k of the ionization process. We note that in this case the $W(\omega)$ relationship functionally has the same character as in

the case of multiphoton excitation of an atom with narrow-spectrum radiation, in which the shift of the atomic levels dominates (Sec. 5, Fig. 8).

The above discussion implies a possibility in principle of determining the distribution $P(F)$ from data on the distribution $W(\omega)$.[34,52] Essentially, this possibility arises from the fact that the perturbation is governed by the shift of the levels.[27]

c) $k_1 > 2, k_2 = 1$. The perturbation of the resonance state is determined both by the shift of the levels and by the ionization broadening of this state, since both processes are proportional to the square of the field intensity. As is well known, apart from the frequencies at which the shift is zero, the real component of the polarizability is numerically larger than the imaginary component.[25] Therefore, one can assume in this case on numerical grounds that the shift dominates. Correspondingly, the $W(\omega)$ relationship is analogous to the case in which the shift dominates. However, it is smoothed out by the ionization broadening of the resonance state. The results of numerical calculations for a number of concrete relationships between δE and Γ_i and for a number of concrete values of k are given in Ref. 51.

The converse situation has also been treated in which one can neglect the shift compared with the ionization broadening of the resonance state.[51,53] Here the $W(\omega)$ relationship has a symmetrical form with a half-width of the order of the width of the resonance level, while we have $W \sim \langle F \rangle^{k-1}$. A practical realization of this situation is improbable. We should bear in mind that one can in no way assume the level shift to be zero at the frequencies at which the first (quadratic) term of the expansion of the polarizability in terms of the field intensity is zero. Here we must pay attention to the second term of the expansion (the hyperpolarizability), which can attain a value comparable with the first term in a number of cases, even when $\mathscr{E} \ll \mathscr{E}_{at}$.[25]

The analysis performed above has assumed that one can employ the concept of the probability per unit time to describe the transition from the excited state to the continuum spectrum. That is, we have not allowed for the possible saturation of this transition. Thus we have been treating only the case in which the relationship holds that $\int_0^\tau W_{iE} dt \ll 1$, where τ is the duration of action of the field. Yet when saturation arises, i.e., the transition from the resonance state to the continuous spectrum occurs with a total probability of unity, the character of the resonance ionization process is determined only by the process of excitation of the resonance state.[48] In each of the treated cases, this leads to obvious changes in the relationships that were presented. The onset of saturation plays an essential role from the practical standpoint. Actually, as we have said above, a typical regime of operation of a laser whose radiation is used to realize multiphoton transitions, is the regime of Q-switching of the resonator, for which a radiation pulse duration $\tau \sim 10^{-8}$ s is characteristic. Correspondingly, saturation sets in at an ionization probability $W_{iE} \sim 10^8$ s^{-1}, or in other words, at a width of the resonance state of $\Gamma_i \sim 10^{-3}$ cm^{-1}. That is, it arises at a broadening on about the same scale as the natural width of the atomic levels. Therefore, as a rule, the case of $\Delta\omega < \Gamma_i$ is not realized.

A dependence of the $W(\langle F \rangle)$ relationship on the detuning of the resonance has been repeatedly observed experimentally.[55,56] Although the overwhelming majority of such experiments has been performed in the radiation field of multifrequency lasers, it is hard to compare the results quantitatively with those of theoretical calculations because one must take into account the space-time inhomogeneities of the radiation intensity in the region of formation of the ions. Reference 57 is an example of taking time inhomogeneity into account. Yet the results of the experiments qualitatively agree well with the conclusions of the theory.

Finally, we must note that generally the narrow-spectrum case can be realized only with a spectral width of the laser radiation not exceeding several cm^{-1}. The point is that, although level shifts of tens and hundreds of cm^{-1} can be realized at fields $\mathscr{E} < \mathscr{E}_{at}$, nevertheless, generally the two-level model of the atom that underlies the analysis being conducted will break down for such values of the shifts.

2) *Resonance ionization by broad-spectrum radiation.* The criterion (6.9), which defines the condition for realization of the broad-spectrum case, implies that the perturbation of the resonance state is small in comparison with the spectral width of the laser radiation. Thus the perturbation does not affect the ionization probability. We recall that the magnitude of the perturbation of the resonance state that enters into this criterion is determined by the averaged characteristics of the nonmonocromatic field in (6.11)–(6.13). The perturbation of the resonance state, which was the cause of the deviations from a power-function relationship in the case of a monochromatic field, plays no role in this case. Therefore the dependence of the ionization probability on the intensity of broad-spectrum radiation has the power-function character $W \sim \langle F \rangle^k$. The probability of resonance ionization in the case of broad-spectrum radiation is qualitatively described by a formula of the form of (6.5), in which the width Γ of the resonance is determined by the effective spectrum of the radiation of order k_1.

Let us examine how the process of resonance ionization by broad-spectrum radiation differs from that by monochromatic radiation. As before, we shall assume in this comparison that the intensity F of the monochromatic radiation equals the mean intensity $\langle F \rangle$ of the nonmonochromatic radiation.

First, the power-function character of the dependence of the ionization probability on the radiation intensity is realized in the nonmonochromatic field up to higher values of $\langle F \rangle$ than the corresponding values of F in the case of the monochromatic field.[49] An analogous effect occurs also for that one of two nonmonochromatic fields that has the larger spectral width. The reason for this effect is rather obvious—the breakdown of the power-function relationship arises from the perturbation of the resonance state, while the latter is manifested at a

greater radiation intensity as the width of the resonance increases. In a sufficiently weak monochromatic field, the width of the resonance is determined by the natural width of the resonance state, while in a nonmonochromatic field in the case of broad-spectrum radiation, it is determined by the effective k_1-order spectrum $S_{k_1}(\omega)$: the width $\Delta\omega_{k_1}$ of the latter is always greater than the spectral width $\Delta\omega$. In particular, when the fluctuations of $\mathscr{E}(t)$ and the shape of the spectrum $F(\omega)$ are Gaussian in type, the width of the effective spectrum is $\sqrt{k_1}\,\Delta\omega$.

Second, in the region of variation of the radiation intensity in which the ionization process in the monochromatic and nonmonochromatic fields is of a power-function type, the ionization probability in the nonmonochromatic field can be either greater or smaller than in the monochromatic field for equal radiation intensities $\langle F\rangle$ and F. In particular, other conditions being equal, the probability of ionization in the nonmonochromatic radiation field of two different lasers is inversely proportional to the width of the laser radiation spectra.

In a number of experiments a resonance ionization process has been observed under conditions of a broad laser radiation spectrum. However, one can extract quantitative data only from the results of Refs. 31 and 58, since the width of the radiation spectrum was controlled therein.

A process was observed in Ref. 58 of three-photon ionization of the sodium atom in the presence of an intermediate two-photon resonance arising under the action of radiation having two different spectral widths. Figure 16 shows the experimental data on the dependence of the ion yield on the radiation intensity. These relationships are of a power-function type with an exponent of $k=3$, which corresponds to the conclusions drawn above. For the radiation having the greater spectral width, deviations from the power-function relationship arise at a greater mean radiation intensity. This corresponds to the conditions for realization of the broad-spectrum criterion (6.9) in the presence of a perturbation of the resonance state. The ion yield at a fixed mean radiation intensity is inversely proportional to the spectral width of the radiation.

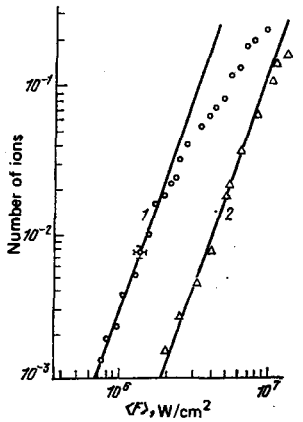

FIG. 16. Experimental values of the ion yield of a process of three-photon ionization of an atom having a two-photon resonance that arises under the action of the radiation from a multifrequency laser at different spectral widths.[58] 1—$\Delta\nu \approx 0.09$ cm^{-1}, 2—$\Delta\nu \approx 1.25$ cm^{-1}. Straight lines—approximation of the experimental data by a $W \sim \langle F\rangle^3$ relationship.

The same process of three-photon ionization of the sodium atom with a two-photon resonance has been observed in Ref. 31. There a study was made of the resonance relationship of the ionization probability $W(\omega)$ to the radiation of a multifrequency laser. The measurements showed that the $W(\omega)$ relationship corresponds to an effective second-order spectrum: the form of the resonance is close to a Gaussian curve, while its width of 0.12 cm^{-1} is about $\sqrt{2}$ times as great as the width 0.08 cm^{-1} of the laser radiation spectrum, which was measured independently. Here the ion yield was proportional to the third power of the laser radiation intensity for every value of the detuning.

7. CONCLUSION

In closing the discussion of the problem of the nonlinear interaction of nonmonochromatic laser radiation with atoms, we must note first a series of problems germane to the topic of this review, but not duly reflected in it.

Upon first turning to nonlinear phenomena *per se* for various reasons we have not discussed certain topics. They include, among others, such important phenomena as the nonlinear scattering of laser radiation, including the generation of higher harmonics, as well as the dissociation of molecules in an infrared field. As regards the process of harmonic generation, it is well known that it is of greatest interest in the case of an extended nonlinear medium in which phase relations are established between the light at the fundamental and multiple frequencies so that a considerable fraction of the energy of the incident light is converted into the harmonic. Under such conditions, nonmonochromaticity of the exciting light exerts the strongest influence on the establishment of phase relationships in the medium,[14] so that the treatment of the effect of nonmonochromaticity of the light on the process of harmonic generation in an isolated atom is not an independent problem. The process of dissociation of molecules in an infrared laser radiation field naturally attracts attention, since the dissociation potential is always far greater than the quantum energy of the radiation. Correspondingly, in principle, the dissociation process can be of an extremely multiquantum type. Thus we can expect that effects involving nonmonochromaticity of the radiation will be very significant in this case. However, numerous experimental facts obtained recently clearly show that dissociation actually is not an extremely multiquantum process. The most likely model is the one in which the dissociation process consists of two stages. The first amounts to an excitation of the molecule involving relatively few photons, while the second consists of collecting energy from the external field under conditions where a quasicontinuum of excited states appears.[59] An elucidation of the role of nonmonochromaticity of the radiation in the process of dissociation of molecules is of evident interest. Only the first steps[60,61] have been taken in this direction, though not

in connection with extreme multiquantum transitions.

The review has not discussed the effect of nonmonochromaticity of the laser radiation on the quasienergy spectrum of a two-level system arising in a strong field.[62] References 19, 34, and 63–65 have derived an analytical solution of this problem for systems having constant dipole moments. In practice, these systems are realized in a large number of cases. These are the hydrogen atom and hydrogen-like states of complex atoms, molecular systems lacking a center of inversion, and systems of spin 1/2 in a constant plus a collinear alternating magnetic field. The results obtained in the cited studies for various special cases indicate the contrast in the action of monochromatic and nonmonochromatic fields. However, this set of phenomena is important in the action of a low-frequency field. Thus it stands apart from the main topic of this review: an atom in a strong nonmonochromatic field in the optical frequency range. (We note that, as applied to molecules having large constant dipole moments in the ground or excited state, the effects that arise can be important also in the optical frequency range.)

Upon turning now to laser radiation, we find two important special cases that have not been studied in detail: single-frequency radiation and multifrequency radiation with phase-synchronized modes. The radiation of mode-synchronized lasers has certain advantages, owing to its extremely high intensity and extremely small pulse duration. Currently it is widely applied for research, including nonlinear-optics phenomena. The justification for our ignoring this multifrequency regime is the determinate character of the radiation with phase-synchronized modes. A description of its interaction with an atom reduces to taking into account the envelope of the pulse. As an example, we can point out the solution of this problem in the case of resonance ionization.[48]

In a number of cases, the action of single-frequency laser radiation is equivalent to that of monochromatic radiation. However, single-frequency radiation is always characterized by a finite spectral width. Hence it also has features specific for nonmonochromatic radiation. In a continuous-wave generation regime, the width of the spectrum of single-frequency radiation is determined by the random phase variation. This radiation is described by the phase "diffusion" model well known in the scientific literature. We note that this model does not take into account fluctuations of the intensity of the radiation. Thus it does not reflect the fundamental properties of multifrequency laser radiation, and hence we have not treated it.

If finally we turn to the measured quantities, we should note that the study of the fluctuations of nonlinear signals, which, as we have said above, has practically not been carried out, is a promising line of study. Thus, for example, in principle one can extract from data on fluctuations of the yield of a k-photon process information on the moments of the intensity distribution of orders higher than k.[66]

Finally we note that we have restricted the discussion to effects that occur under conditions in which the concept of the probability per unit time can be applied. Taking into account saturation and a derivation of the relationships for the total probabilities substantially complicates the description of the elementary nonlinear-optical phenomena in a nonmonochromatic field.[47]

In closing we must note that the set of problems that we have discussed concerning the role played by the nonmonochromaticity of the laser radiation field in exciting elementary nonlinear-optical phenomena is not only of general physical interest, but also has great significance in practice. Actually, for a broad set of applications involving the selective action of laser radiation on atoms and molecules, the conditions of experiment in which the interaction occurs with an isolated atom are optimal. As regards multifrequency lasers, they make it possible to obtain a considerably larger radiation intensity than single-frequency lasers do, so that the promise that they offer in practical applications is undoubted. Finally, the elucidation of the physical essense of nonlinear-optical phenomena that arise in an extended medium requires a knowledge of the details of the character of the nonlinear interaction with an isolated atom.

We wish to call the readers' attention to the fact that the bibliography given in this review does not fully reflect the publications, since, as a rule, we have granted preference to monographs and reviews, as compared with original papers.

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