mined by formulas such as (4.2)–(4.6), where it is necessary to replace the frequency $n\omega$ by $(n\pm1)\omega$ and the matrix element W_{01} by the composite second-order matrix element $\tilde{W}_{01}$. The spectral characteristics of the stimulated absorption do not differ in this case from those examined above, and the absorbed (emitted) energy is lower than in the vicinity of the resonant frequency by a factor $(F_0/F_{at})^2$. When $\Omega\sim\omega$, the dissipated energy is determined by an expression of the form

$$\Delta\mathscr{E}=\omega\int dE\left\{\left|\int dE'\int_0^t d\tau\, a_{E'}^{0*}(a_E^{0*}a_{E'}^{0}\tilde{W}_{00}+a_E^{1*}a_{E'}^{1}\tilde{W}_{11})\right.\right.$$
$$\times\exp[i(E-E'+\omega-\Omega)\tau]\Big|^2-\Big|\int dE'\int_0^t d\tau\, a_{E'}^{0*}(a_E^{0*}a_{E'}^{0}\tilde{W}_{00}$$
$$\left.\left.+a_E^{1*}a_{E'}^{1}\tilde{W}_{11})\exp[i(E-E'-\omega+\Omega)\tau]\right|^2\right\},\qquad(4.8)$$

where $\tilde{W}_{00}$ and $\tilde{W}_{11}$ are the diagonal composite matrix elements.

In the limiting cases $\Gamma_i\ll\Gamma_f$ and $\Gamma_i\gg\Gamma_f$ this expression yields respectively

$$(\Delta\mathscr{E})_\omega=\frac{\omega\varepsilon}{R_i}(\tilde{W}_{00}-\tilde{W}_{11})^2\sum_{\pm}\frac{(\pm1)}{(\Omega-\omega\mp R_i)^2+\Gamma_i^2/4},\quad\Gamma_f\gg\Gamma_i,\qquad(4.9)$$

$$(\Delta\mathscr{E})_\omega=4\frac{\tilde{W}_{00}\tilde{W}_{11}\varepsilon\Omega(\omega-\Omega)}{R_i^2[(\Omega-\omega)^2+(\Gamma_i\Gamma_f^2/16R_i^2)^2]},\quad\Gamma_i\gg\Gamma_f.\qquad(4.10)$$

Comparison of these expressions with Eqs. (4.4) and (4.6) shows that in both cases $(\Delta\mathscr{E})_\omega\sim(F_0/F_{at})^2(\Delta\mathscr{E})_{n\omega}\ll(\Delta\mathscr{E})_{n\omega}$. As applied to the case $\Gamma_i\gg\Gamma_f$, this conclusion differs from the corresponding result for spontaneous emission. This is due to the cancellation, in stimulated transitions, of the fundamental process responsible for the spontaneous emission by atoms at frequencies near the pump frequency ω.

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Multiphoton processes in the radiation field of a multimode laser

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The probabilities of multiphoton ionization by single-mode and multimode laser pulses of given mean field strength have been measured. For a power-law five-photon process, the probability in the field of the multimode laser is greater by a factor of 5! For an eleven-photon process, the corresponding factor has been found experimentally to be much less than 11! This reduction is explained by a departure from the power-law variation in the region approaching tunneling.

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§1. INTRODUCTION

One of the main problems in studying elementary nonlinear optical processes is the determination of the absolute probabilities of bound-bound and bound-free transitions.[1,2] For technical reasons, the experimental data are usually more readily obtained by using the radiation from multimode lasers. For a given mean intensity in a radiation pulse, the transition probability is determined essentially by the mode content of the laser

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radiation. When the transition is highly nonlinear, the probability in the field of a multimode laser may exceed by several orders of magnitude the probability in the field of the single-mode radiation for the same mean intensity W_0.

A well-known example is the radiation field emitted by a thermal source and a power-law nonlinear process for which the probability ratio is

$$W_{\text{therm}}/W_0 = G_{k_0} = k_0!,$$

where k_0 is the number of absorbed photons and the statistical factor G_{k0} is the ratio of the corresponding higher correlation functions. It is known that the mode content of the radiation from a powerful solid-state laser operated under giant-pulse conditions is very dependent both on the laser design and on the operating conditions. From the point of view of using the laser radiation for studying nonlinear optical phenomena, it is clearly valuable to operate the laser so that the properties of its multimode radiation are as close as possible to the properties of the radiation emitted by the thermal source. When this can be done, it is possible to reproduce the results in successive experiments, and to compare the results obtained in the field of a multimode laser with the theoretical calculations which, as usual, are performed on the assumption that the radiation is perfectly monochromatic. It is then necessary for the laser to emit a sufficient number of modes with a random distribution of amplitudes and phases.

From our point of view, the only case of acceptable multimode operation of a powerful neodymium laser with a random distribution of amplitudes and phases of the generated modes is reported in[3]. To ensure that the radiation field of this kind of multimode laser is equivalent to the radiation field of a thermal source, the number of generated modes must increase with the number of photons involved in the nonlinear process under investigation.[4-6] Experimental data[6] have clearly shown that the statistical factor G_{k0} depends on the number of generated modes and their degree of locking. However, there has been no direct experimental demonstration of the fact that, when the number of random modes and the degree of nonlinearity of the process k_0 are large enough, the statistical factor is given by

$$G_{k_0} = k_0! \qquad (1)$$

All the available experiments were performed for processes with $k_0 = 2$ or 3.

The results of an experiment corresponding to $k_0 = 5$ are given in Sec. 3 of the present paper. Sections 4 and 5 are concerned with the elucidation of the restrictions imposed on (1) by the high degree of nonlinearity of the process and hence the very high field strength for which the process was observed.

The same phenomena that lead to the dependence of the output of the nonlinear process on the number of modes and on mode locking are also responsible for fluctuations along a series of emitted laser pulses with fixed mean intensity per pulse. For a random distribution of phases, the variance of the output is determined by the number of modes and the degree of nonlinearity of the process, and may exceed the mean output.[7] We have selected for investigation the direct multiphoton ionization of atoms and the field of a neodymium glass laser producing giant pulses. The probabilities of multiphoton ionization in the field of single-mode and multimode lasers was compared for a given measured mean intensity per pulse.

§2. FORMULATION OF EXPERIMENT

A standard experimental arrangement was used to observe the multiphoton ionization of atoms (see, for example,[1]). Light from the laser was used to illuminate a target consisting of neutral atoms. Ions produced in the target and the intensity of the radiation in the region in which the ions were produced were recorded.

A. Laser radiation. We used two neodymium glass lasers producing giant pulses. Both lasers generated a single simple transverse mode, and this was achieved by the standard method of inserting a diaphragm into the resonating cavity. One of the lasers produced one longitudinal mode; use was made of a dye for passive Q-switching and a composite resonator was employed. The other laser generated many longitudinal waves; here, Q switching was achieved with a rotating prism. Dispersing elements (Fabry–Perot etalons) were introduced into the cavities of both lasers and ensured that the generation frequency could be varied continuously within the limits of about 50 cm^{-1} on either side of the luminescence peak of the neodymium glass (≈ 9440 cm^{-1}). In both lasers, the radiation density was less than the threshold value (approximately 100 MW/cm^2), above which the nonlinear properties of the neodymium glass became appreciable.[3] Amplifying stages operating under linear conditions were used to produce the high intensity necessary for the experiments.

Particular attention was paid to the control of the mode content of the laser radiation from both lasers. The spectrum of the multimode laser radiation was measured with a diffraction grating spectrograph having a dispersion of about 1 cm^{-1} per millimeter. The spectrum of the multimode laser radiation had a bell-shaped form with a half-width of about 10 cm^{-1} which, for the particular geometry of the cavity, corresponded to about 4000 longitudinal modes. Single-mode generation control was achieved with the aid of the same spectrograph and a Fabry–Perot interferometer with a resolution of 0.01 cm^{-1}, and by recording the time distribution of the radiation with a resolution down to 10^{-9} sec. When the mode separation was less than 0.01 cm^{-1}, beats between the modes distorted the smooth time distribution corresponding to the single-mode generation. The criteria for single-mode generation were the presence of a single line recorded by spectrograph and interferometer, and a smooth bell-shaped time distribution. Single-mode generation control was carried out for each laser pulse. When the laser was properly tuned, the single-mode generation could be achieved for more than 90 out of 100 successive giant pulses. The radiation from both lasers was accurately plane-polarized.

B. Measurement of ionization probability. If we suppose that the ionization probability W is related to the radiation intensity F by the power-law formula

$$W=\alpha_{k_0}F^{k_0}, \tag{2}$$

where α_{k0} is the cross section for direct ionization connected with the absorption of $k_0=\langle I/\hbar\omega+1\rangle$ photons, the amplitude of the ion signal is given by[11]

$$A_i=\beta n_0 W\tau_{k_0}V_{k_0}=\beta n_0\alpha_{k_0}F^{k_0}\tau_{k_0}V_{k_0}. \tag{3}$$

In this expression, β is the sensitivity of the ion recording equipment, n_0 is the density of neutral atoms, F is the radiation intensity in the region in which the ions are produced, and

$$\tau_{k_0}=\int\psi^{k_0}(t)\,dt; \qquad V_{k_0}=\int\varphi^{k_0}(x,y,z)\,dv$$

are, respectively, the effective pulse length and target volume for the process connected with the absorption of k_0 photons.

The radiation intensity is given by

$$F=Q/s\tau,$$

where Q is the energy in the radiation pulse,

$$s=\int\eta(x,y)\,ds, \qquad \tau=\int_0^T\psi(t)\,dt$$

are, respectively, the area of the focal spot and the length of the laser pulse normalized to the maximum of the functions η and ψ, and T is the length of the giant pulse over whose envelope the integration is carried out.

The space-time distributions of radiation from the two lasers were different because of differences in the geometry of the resonating cavities and the operating conditions. The values of ψ, η, and φ were therefore measured in each experiment. Since the target was illuminated simultaneously by both lasers, it was assumed that the quantities β and n_0 remained constant during the measurement process. Since the same equipment was used for measuring A_i, Q, ψ, η, and φ, only random measurement errors had to be taken into account, and this substantially increased the precision of the experiment.

It is important to note that, when an atomic medium is ionized by multimode laser radiation, the ion signal will fluctuate even when the energy of the radiation, the field distribution in space, and the time envelope of the giant pulse remain constant along a series of successive pulses. Changes in the ion signal will occur under these conditions because the time fine structure of the giant pulse, which is usually smoothed out when the radiation is recorded with a photodiode and an oscillograph, cannot be reproduced exactly. The spread in the ion signal can be estimated with the aid of the probability density function $p(F/\overline{F}_T)$ for the intensity of the multimode laser radiation, where $\overline{F}_T$ is the mean intensity in a time T (the axial period of the radiation).

This function has a characteristic feature: it vanishes when $F/\overline{F}_T$ is greater than the number N of longitudinal modes. This behavior of $p(F/\overline{F}_T)$ is connected with the fact that there are only N independent values of instantaneous intensity during the axial period (according to the number of correlation intervals that can be fitted into the axial period), and the maximum difference between F and $\overline{F}_T$ is realized when the entire energy of the axial period is localized in the shortest time interval, i.e., all the values except one are zero. It is possible to obtain an explicit expression for $p(F/\overline{F}_T)$ by equating $\overline{F}_T$ to the average over N independent intensity values. Assuming that the laser radiation field follows a Gaussian distribution, and using the well-known v^2 distribution for the ratio of the sums of normal quantities, we obtain

$$p\left(\frac{F}{\overline{F}_T}\right)=\begin{cases}\dfrac{(N-1)\,(N-F/\overline{F}_T)^{N-2}}{N^{N-1}}, & 0\leqslant\dfrac{F}{\overline{F}_T}\leqslant N\\ 0 & \text{in the remaining cases}.\end{cases}$$

The probability density $p(F/\overline{F}_T)$ can be used to calculate the dependence of the mean normalized ion signal on the degree k_0 of the nonlinearity of the process and the number N of longitudinal modes:

$$G_{k_0}=\int_0^N\left(\frac{F}{\overline{F}_T}\right)^{k_0}p\left(\frac{F}{\overline{F}_T}\right)d\left(\frac{F}{\overline{F}_T}\right)=\frac{k_0!N^{k_0}(N-1)!}{(N+k_0-1)!}. \tag{4}$$

We note that an analogous relationship was obtained in[6] by a different method. The function $p(F/\overline{F})$ can also be used to calculate the variance of the normalized ion signals:

$$\sigma_{k_0}^2=\frac{1}{N}\left[\int_0^N\left(\frac{F}{\overline{F}_T}\right)^{2k_0}p\left(\frac{F}{\overline{F}_T}\right)d\left(\frac{F}{\overline{F}_T}\right)-\left(\int_0^N\left(\frac{F}{\overline{F}_T}\right)^{k_0}p\left(\frac{F}{\overline{F}_T}\right)d\left(\frac{F}{\overline{F}_T}\right)\right)^2\right]=\frac{G_{2k_0}-(G_{k_0})^2}{N}. \tag{5}$$

It is clear from (4) and (5) that, for given k_0 and N, the variance may exceed the average which, in principle, is not unexpected for nonlinear processes. Thus, for $N\sim1000$ and $\sigma_5/G_5\sim0.3$, we have $\sigma_{11}/G_{11}\sim10$. This last result shows that measurements corresponding to $k_0=11$ may encounter difficulties connected with the large spread of the ion signals. To ensure the necessary precision of measurement, the receiving apparatus must have a sufficient dynamic range, and the number of laser pulses must be sufficiently large. In the above example, the measurement error for $k_0=11$ is comparable with the mean when the number of laser pulses is ~100.

Unfortunately, the dynamic range of the apparatus used to measure the amplitude of the ion signal was too low (~1000) and, therefore, the signal amplitude could not be directly measured for a fixed mean intensity of single-mode and multimode laser radiation. The experiment was, therefore, designed to measure the radiation intensities for which the ion-signal amplitudes were equal. These data can be used together with the data on the function $W(F)$ to calculate the factor G.

Since the results of our investigation were quantitative

in character, particular attention was paid to their reproducibility, and different control experiments were also performed. The quantities A_i and F were measured by a method well established for measurements of multiphoton cross sections (see[1,8]).

§3. FIVE-PHOTON POWER-LAW PROCESS

We recorded the five-photon direct ionization of the sodium atom. Previous experiments[8] gave exhaustive information on the five-photon ionization of sodium by neodymium-glass laser radiation. The ionization is observed for field strengths of $\mathscr{E} \approx 10^6$ V/cm and adiabatic parameter $\gamma = T_{tun}/T \approx 5 \times 10^2$, where T_{tun} is the electron tunneling time and T is the period of the external field. The condition $\gamma \gg 1$ corresponds to the power law given by (2) for the ionization probability as a function of radiation intensity with $k_0 = 5$. This value is satisfactorily confirmed by experiment.[8] When the neodymium-glass laser radiation is linearly polarized, we have direct ionization: the initial detuning between the energy $k < k_0$ of photons with frequency $\omega = 9440$ cm^{-1} and the energies of transitions between the bound states is large and does not vary very much with the field. In accordance with the data in the literature,[4-6] the number of modes with a random phase distribution in the case of the five-photon process should be greater than 50 for the realization $G = 5!$ As indicated above, our multimode laser emitted 4000 modes. The five-photon process which we have chosen was therefore a perfect detector for measuring the function G_5.

Figure 1 shows the results obtained in the field of a single-mode and multimode laser, indicating the precision of the relative measurements of the ion-signal amplitudes and the radiation intensity. When the ion signal is not saturated, i.e.,

$$\int_0^T W\,dt \ll 1,$$

where T is the pulse length, the experimental data can be approximated to by a power law. The quantity $k = \partial \log A_i / \partial \log Q$ is equal to 4.95 ± 0.05 in the case of the single-mode laser and 4.8 ± 0.2 in the case of the multimode laser, i.e., to within experimental error $k = 5 = k_0$.

It is quite clear that a constant signal amplitude (constant ionization probability) is achieved for relatively high mean radiation intensity of the single-mode laser as compared with the mean intensity of the multimode laser: $F_0/F_m = 2.56 \pm 0.25$. Analysis of the experimental results, assuming the validity of the power law (2) with $k_0 = 5$, yields the following ratio of ionization probabilities for fixed multimode and single-mode radiation intensities (W_m and W_0)[9]:

$$W_m/W_0 = 10^{2,04 \pm 0,2} = G_5.$$

The measured value of G_5 is equal to $5! = 10^{2.08}$ to within the experimental error. The spread of the ion signals, referred to a common intensity, is shown in Fig. 2a. The result $\sigma_5/G_5 \approx 0.25$ is in good agreement with estimates based on (4).

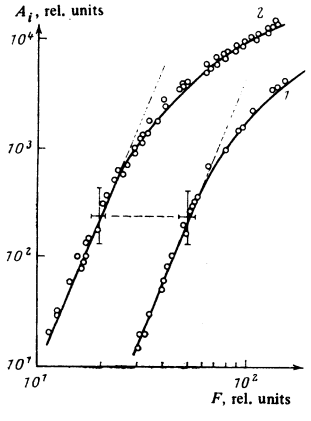

FIG. 1. Number of sodium ions as a function of the pulse-averaged intensity of single-mode (1) and multimode (2) laser radiation. The relative precision with which A_i and F were measured is indicated.

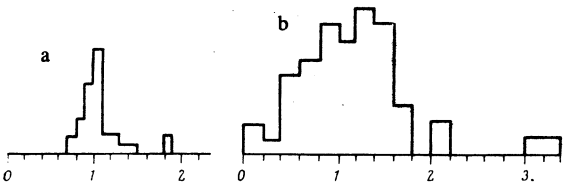

FIG. 2. Spread of the ion signal at constant laser radiation intensity: a) ionization of the sodium atom; b) ionization of the xenon atom.

Summarizing the results of this experiment, we note that the data obtained for $k_0 = 5$, taken together with the previous results for $k_0 = 2$ and 3, cover practically the entire range of nonlinearity that is of interest in practice. There is also the fundamental point that, if we remain in the optical frequency band, the observation of processes with a greater degree of nonlinearity requires much greater field strengths, which leads to a reduction in the adiabatic parameter to values for which the condition $\gamma \gg 1$ ceases to be satisfied.[1] As noted in the Introduction, one would then expect a reduction in the difference between the output of nonlinear processes in the single-mode and multimode radiation fields. Our experiment on the ionization of the xenon atom was concerned with this case.

§4. IONIZATION AT $\gamma = 5$

The ionization of the xenon atom by neodymium-glass laser radiation was investigated in detail in[1,10]. Ionization is observed when the field strength is $\sim 5 \times 10^7$ V/cm, which corresponds to adiabatic parameter $\gamma \approx 5$. Experiment[10] shows that the spectrum of the xenon atom is highly disturbed by the field, but it is possible to choose a radiation frequency for which the ionization process is direct, i.e., there are no intermediate resonances with bound electronic states. When the experiment on the ionization probability is performed in the field of two lasers, namely, single-mode and multimode lasers, there is the additional problem of finding the radiation frequency at which, in both cases, the ionization process has a direct character. Existing experimental data could not be used for this purpose because the xenon spectrum was too highly perturbed and the precision of absolute determinations of the radiation in-

tensity was not high enough. The frequency dependence of the ionization process was also investigated by the method described in[10], in which the amplitude of the ion signal was measured as a function of the radiation frequency for a fixed radiation intensity. In both cases, there is a region in which the signal amplitude is independent of frequency, and this is used as a criterion for the absence of intermediate resonances. It is important to note two facts: firstly, the data obtained for the multimode laser are in good agreement with the data for the analogous laser reported previously in[10]. At the same time, the data for the single-mode laser are substantially different, which is probably connected with a difference between the field strengths in which the ionization was observed in the two cases, and hence with different perturbation of the atomic spectrum. Secondly, there is a frequency band at 9410 cm^{-1}, in which the ionization process has a direct character in both cases. The ionization probabilities in the fields of the two lasers were therefore measured at the frequency of 9410 cm^{-1}.

If we suppose that the ionization process can be described by a power-law formula with exponent $k_0 = 11$, then existing data[4-6] show that about 1000 random modes are necessary to ensure that $G_{11} = 11!$ Our multimode laser radiated 4000 modes. The results of our measurements are shown in Figs. 3 and 2b. As in the case of sodium, it is clear that, to produce fixed ionization probability in single-mode radiation, the pulse-averaged intensity must be much greater than for the multimode radiation: $F_0/F_m = 2.85 \pm 0.5$. In the absence of saturation, $k = 10.8 \pm 0.8$ for single-mode radiation and $k = 10.7 \pm 0.7$ for multimode radiation.

It is also clear that the relative spread of the ion signals, i.e., $\sigma_{11}/G_{11} \approx 0.65$, is greater than for $k_0 = 5$ although it is much smaller than the spread corresponding to (4). One of the reasons for this difference between the calculated and experimental data may be the small dynamic range of our apparatus (~100). A further reason may be the fact that we did not record cases for which the amplitude of the ion signal lay outside the dynamic range, or signals for which the energy in the radiation pulse measured by the monitoring calorimeter deviated from the mean by more than a given amount (~5%). This method of measurement automatically reduces the number of large deviations. Assuming that $k_0 = 11$, we obtain the following ratio at constant mean intensity[11]:

$$W_m/W_0 = 10^{3\pm1} = G(\gamma \approx 5).$$

The measured $G(\gamma \approx 5)$ is much less than $G_{11} = 11! = 10^{7.6}$.

In principle, there are three possible methodological reasons for this difference. They are: the presence of uncontrollable errors, partial mode locking, [6] and insufficient number of generated modes. The first two of these must be excluded because our experiment was performed under conditions identical with those used in the experiments with sodium, in which the magnitude of G was in good agreement with $G_5 = 5!$ Although the number of generated modes was such that we did not have an

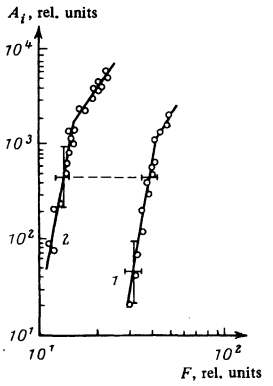

FIG. 3. Number of xenon atoms as a function of the pulse-averaged intensity of single-mode (1) and multimode (2) laser radiation. The relative precision of the measurements is indicated.

appreciable margin in the experiment with xenon, as compared with the theoretically required magnitude, the data obtained by calculation in[4,5] and by experiment in[6] show that the observed value of G could be realized only when the number of modes was ~10, which was so much smaller than the 4000 actually realized in our experiments that this particular factor could not play an important role. We are therefore forced to assume that there are departures from the power-law form of $W(F)$ due to the fact that the adiabatic parameter is not large enough ($\gamma \approx 5$ is closer to $\gamma \sim 1$ than to $\gamma \gg 1$).

§5. DEPENDENCE OF THE STATISTICAL FACTOR ON THE ADIABATIC PARAMETER

It follows from the Keldysh theory[12] that the power-law formula (2) relating the ionization probability and radiation intensity is valid only for $\gamma \gg 1$. In the other limiting case, when $\gamma \ll 1$, the function $W(F)$ is an exponential, typical for the tunnel effect. In the intermediate region in which we are interested, in which $\gamma \sim 1$, the function $W(F)$ deviates from the power-law dependence and varies much more slowly. This may be looked upon as an indication that, as γ approaches unity, the statistical factor should decrease.

To calculate the statistical factor as a function of γ, we use the expression for the ionization probability as a function of the adiabatic parameter, obtained theoretically in[12,13] in the quasiclassical (adiabatic) approximation:

$$W(F) = D(\gamma)\exp\{-2If(\gamma)/\omega\}S(\gamma, \omega). \quad (6)$$

The functions $D(\gamma)$ and $S(\gamma, \omega)$ in (6) are slowly-varying functions of γ as compared with the exponential function, so that we took into account only the exponential term. The probability (6) was averaged over the statistical ensemble with a Gaussian weight function

$$\rho(F, \bar{F}) = \frac{1}{\bar{F}}\exp\left(-\frac{F}{\bar{F}}\right), \quad (7)$$

where the mean is given by

$$\bar{F} = \int_0^\infty F\rho(F, \bar{F})\,dF.$$

The ionization probability in the field of the multimode

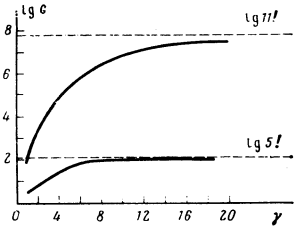

FIG. 4. The function $G(\gamma)$ corresponding to (8). The calculation was carried out for the two cases $k_0=11$ and $k_0=5$.

laser is, therefore, given by

$$W_m(\bar{F})=\int_0^\infty W(F)\rho(F,\bar{F})\,dF. \tag{8}$$

It is readily seen from (7) and (8) that, when $\gamma\gg 1$, for which $W_m(\bar{F})\sim F^{k_0}$, we have $G=W_m(\bar{F})/W_0(F)=k_0!$ If we substitute the explicit form of $f(\gamma)$ from (6) in (8), and carry out the numerical integration, we obtain the function $W_m(\bar{F})/W_0=G(\gamma)$ shown in Fig. 4 for two special cases (W_0 is the ionization probability in the field of the single-mode laser).[14,15]

It is clear from Fig. 4 that, when $\gamma\approx 5$, the statistical factor $G(\gamma)$ corresponding to $k_0=11$ is substantially different from $G_{11}=11!$, and agrees to within an order of magnitude with the result obtained in the experiment with xenon, discussed in the previous section. Calculations thus confirm that the observed deviation of the statistical factor from 11! is a consequence of the intermediate character of the ionization process for $\gamma=5$ and, in particular, of the fact that the power-law formula given by (2) is not accurate enough to describe the probability of ionization as a function of light intensity.

Integration shows that the main contribution to (8) for $\gamma\gtrsim 1$ is due to the field region $F\approx k_0\bar{F}$. This enables us to carry out a number of simple estimates. Thus, we can readily estimate the precision with which (6), valid for a short-range potential, is also valid for the ionization of a real atom. Using published data[13] for $\gamma<\gamma_c=(\mathscr{E}_{at}/\mathscr{E})^{1/2}$ (it is clear that this is satisfied in our case), we find that the factor representing the long-range Coulomb potential is $f_c\sim 1/F$ and provides the factor $\sim 1/k_0$ in $G(\gamma)$. Although this reduces the effect, the reduction is small for $\gamma=5$, i.e., in the case in which we are interested.

There is no doubt that a more accurate calculation of $G(\gamma)$, including the effect of polarization, is necessary. Such data are essential, in particular, for the analysis of the polarization dependence of ionization probability measured for $\gamma\sim 1$ in the radiation field of a multimode laser. However, even the estimates made above indicate the overall character of the function $G(\gamma)$.

We note that the calculations of $G(\gamma)$, shown in Fig. 4, do not answer the question as to what are the conditions for the nonlinear process to become independent of the mode content of the laser radiation. The point is that, if we remain in the optical frequency band, data obtained for $\gamma\ll 1$ are meaningless because the condition $\mathscr{E}\ll\mathscr{E}_{at}$ is violated in this region and the original theoretical relationships for $W(\gamma)$[12,13] are no longer valid. However, simple analysis of the general formula for $W(\gamma)$, given by Keldysh[12] (see, for example,[1]), will show that, even for $\gamma<0.1$, the ionization probability is practically independent of the electromagnetic field strength, and ionization occurs in a time comparable with the characteristic atomic time. Clearly, under such conditions, the ionization probability will also be independent of the mode composition of the laser radiation.

At first sight, it might appear that the conclusion that there is an appreciable deviation from the power-law process in the ionization of xenon at $\gamma=5$ is in conflict with the form of the function $W(F)$ observed in the same experiments and approximated to by an expression of the form $W\sim F$.[11] However, there is, in fact, no contradiction. The power-law approximation over a small interval corresponding to $\gamma\approx 5$, using $W(F)$ calculated from the formula taken from[12,13], and from the exact formula for circularly polarized light,[16] yields $k=10.6$ (see[10]), and this agrees to within experimental error with $k_0=11$.

§6. CONCLUSIONS

Summarizing the results of our investigations, we may conclude that the statistical factor for power-law processes excited by multimode laser radiation containing a sufficient number of random modes is equal to $k_0!$, where k_0 is the number of photons absorbed in the elementary event. In the case of bound-free transitions (ionization), the statistical factor decreases as the adiabatic parameter approaches unity, and this is connected with the fact that $W(F)$ is a more slowly-varying function in this region as compared with the corresponding power-law dependence. The question of bound-bound transitions (multiphoton resonance) will require a separate experimental investigation since, among other things, theory predicts a number of new effects.[17]

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Features of absorption of intense IR emission by SF_6 molecules

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It is demonstrated experimentally that the frequency of the minimum absorption by SF_6 molecules is shifted into the long-wave region at high intensities of the incident radiation and at high IR pulse durations exceeding the times of rotational and vibrational relaxation. The observed singularities of the absorption spectrum are illustrated with the chemical reaction SF_6+H_2 as an example, and an explanation of the results is presented.

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A study[1] of the selective dissociation of the SF_6 molecule in a strong IR laser field has revealed a frequency shift that brings about a most effective rate of collisionless dissociation of this molecule. We have shown earlier[2] that when the SF_6 molecule is exposed to a strong pulse of CO_2 laser radiation with a pulse duration τ_p exceeding the rotational and vibrational relaxation times ($\tau_{r\text{-}r}$ and $\tau_{v\text{-}v}$), a strong change is observed in the absorption spectrum of the SF_6 molecules when the intensity of the incident radiation is changed. The present paper is devoted to a more detailed study of this phenomenon at various IR intensities.

1. EXPERIMENTAL SETUP

An oscillogram of the irradiating pulse is shown in Fig. 1. The radiation source was a pulsed electric-discharge atmospheric-pressure CO_2 laser similar to that described in[3]. The electrode length was 90 cm, and the distance between electrodes was 5 cm. The laser-radiation frequency selection was effected with a diffraction grating operating in the self-collimation regime. The exit mirror of the laser was an NaCl substrate on which a dielectric coating having a reflection coefficient 40% was sputtered. The working medium was a mixture of the gases CO_2, N_2 and He in a ratio 1 : 1 : 2. The lasing pulse duration (Fig. 1) was ~3 μsec at half-height. The CO_2 laser energy on individual vibrational-rotational transitions reached 6 J.

The cell (Fig. 2) for the investigation of the IR absorption and for the initiation of the chemical reaction in the SF_6+H_2 mixture was 40 cm long. The cell windows were made of NaCl and were inclined at the Brewster angle. The CO_2-laser radiation incident on the cell was collimated into a beam of ~6 mm diameter and practically homogeneous over its cross section (over the entire length of the cell). The waveform of the lasing pulses was registered with a Ge : Au photoresistor (T_{work} = 77 K), the signal from which was fed to the input of an S8-2 oscilloscope with a bandwidth up to 8 MHz. The time constant of the radiation receiver was not worse than 200 nsec. The calorimetric energy meters (of the KIM type) ensured a measurement reproducibility

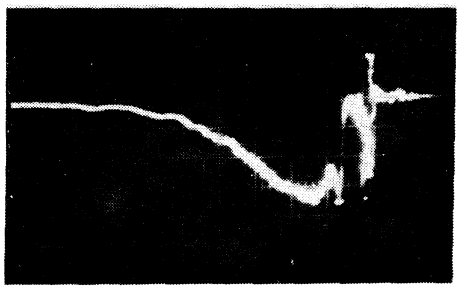

FIG. 1. Oscillogram of CO_2-laser pulse. Time sweep 1 μsec/div.

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