Multiphoton ionization of atoms

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The most important results of study of multiphoton ionization of atoms are reviewed. Discussions are presented of specific features of the process, theoretical methods of description of ionization of atoms in a strong light field, the basic principles of design of experiments in which intense laser radiation is used, results of investigation of direct and resonance processes of multiphoton ionization, and the mutual relation between multiphoton processes (multiphoton excitation and ionization) and various phenomena leading to perturbation of the spectrum of bound states of an electron in an atom under the influence of a strong light field.

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INTRODUCTION

Interaction of an atom with a light quantum–a photon–is one of the most studied areas of the physics of the microworld, which stimulated the development of quantum mechanics at the beginning of the centruy. However, with rare exceptions, all processes investigated up to the present time have been associated with absorption by an atomic electron of one photon. Discussion of one-photon processes determined the universal nature of Einstein's laws and also the red limit of the photoelectric effect. Nevertheless, it has always been clear that, strictly speaking, that there is no red limit, that it is not a fundamental limitation but only a quantitative limitation associated with the low probability of multiphoton processes. The possibility in prinicple of multiphoton transitions was already understood at the time of the development of quantum mechanics. The well known effect of tunneling of an atomic electron through a potential barrier under the action of an external constant field was essentially an adiabatic limit for processes associated with absorption of many photons. Because of the anharmonicity of the spectrum of bound states of an electron in an atom, a multiphoton transition can be described only as a series of virtual transitions, each of which occurs with violation of the conservation of energy. Since in virtual transitions the electron lifetime in the intermediate state is determined not by relaxation but by the energy-time uncertainty relation, it is obviously very small. In order that the electron be able to transfer from this intermediate state to a state with higher energy, it is necessary that the next photon be absorbed after a very short time interval, i.e., a very high light intensity is necessary. Only the creation of high-power lasers has provided experimenters with a source of sufficiently high intensity for observation of multiphoton absorption of light by atoms and thereby stimulated the development of a theoretical description of multiphoton processes.

In what follows we will often use the term "strong light field." This term is evidently not equivalent to the term "high intensity light field," and also there can be no single quantitative criterion as to what field must be considered strong. For example, for a two-level system with relatively low intensity a resonance field can in a certain sense be considered strong–it is well known that a two-level system + resonance field represents a new quantum state whose wave function is a linear combination of the wave functions of the initial states. On the other hand, an "atomic field," i.e., the field intensity in the orbit of an electron in the hydrogen atom in the ground state $\mathscr{E}_{at} \approx 5 \times 10^9$ V/cm evidently can be considered weak in comparison with the limiting field strength $\sim 10^{11}$ V/cm which can be obtained at the present time by using high-power laser radiation.

1. SPECIFIC FEATURES OF THE PROCESS OF MULTIPHOTON IONIZATION OF ATOMS

a) Keldysh's theory. The dependence of the probability of ionization of an atom in a strong light field on the basic parameters characterizing the atom (the ionization potential I) and the field (frequency ω and intensity $\mathscr{E}$) was investigated by Keldysh[1] by solution of the model problem of removal of an electron from a rectangular potential well of depth $I \gg \hbar\omega$. He took it into account that the electron extracted from the well is accelerated by the light field and has an average energy

$$E_{osc} = \frac{e^2\mathscr{E}^2}{4m\omega^2}.$$

We are interested in the limiting cases of the general solution obtained by Keldysh.[1] In one limiting case it turned out that the probability of electron knockout is related to the field strength by the following power law:

$$W = \alpha_{k_0}(I, \omega)\mathscr{E}^{2k_0}, \qquad (1)$$

where $\mathscr{E}^2 = F$ is the intensity of the radiation and k_0 is the number of photons whose absorption is necessary to satisfy conservation of energy:

$$k_0 = \left\langle \frac{\tilde{I}}{\hbar\omega} + 1 \right\rangle, \qquad (2)$$

and $\tilde{I} = I + E_{osc}$ is the effective potential for ionization of the atom in a strong light field. Equation (1) has a

form typical for a multiphoton process—a power-law dependence of probability on radiation intensity; $\alpha_{k_0}(I, \omega)$ is a constant independent of the intensity and which plays the role of the cross section for the multiphoton process.

In the other limiting case, for high field strength or low frequency of radiation, the probability depends exponentially on the field strength:

$$W = \frac{B}{\sqrt{\mathscr{E}}} \exp\left(-\frac{C}{\mathscr{E}}\right), \qquad (3)$$

where B and C are constants. In Eq. (3) the exponential has the well known appearance of the probability of tunneling in a constant field.

The realization of the multiphoton or tunneling nature of the ionization process is determined physically by whether or not the electron can traverse the potential barrier in one period of alternation of the light field. If the time of traversal of the barrier is less than the time in which the phase of the field changes, then the transition has the nature of tunneling. We can assume that the time of tunneling is $\tau = l/v$, where the barrier width $l \sim \sqrt{I}$, and the electron velocity is $v \sim \mathscr{E}$; then the well known adiabaticity parameter, which consists of the ratio of the time of tunneling to the period of the field, has the form

$$\gamma = \frac{\omega}{\omega_{\text{tun}}} = \frac{\omega \sqrt{2mI}}{e\mathscr{E}} = \sqrt{\frac{2m\hbar\omega^3}{e^2}} \sqrt{\frac{k_0}{\mathscr{E}^2}}. \qquad (4)$$

Thus, Eq. (1) should be satisfied if $\gamma \gg 1$, and Eq. (3) if $\gamma \ll 1$. Figure 1 illustrates a specific situation in ionization of the hydrogen atom by visible light and by infrared radiation.

Two factors do not permit us to use the elegant results of Keldysh[1] for quantitative description of the ionization of atoms by a strong light field.[1)] In the first place, it is necessary to take into account the effect of the Coulomb field of the atomic core on the electron ejected from the atom. In the second place, it is necessary to evaluate the contribution of transitions in the spectrum of bound states.

Although Keldysh's theory does not provide a quantitative description of the multiphoton ionization of an atom, its value cannot be overestimated, since it permitted determination of the fundamental laws of the ionization process in a strong light field.[2)]

b) The direct ionization process. It can be seen from Eq. (1) that for $\gamma \gg 1$ the ionization occurs as the result of absorption of k_0 photons by an atomic electron. If the energy $k \cdot \hbar\omega$ (where $1 \leq k < k_0$) differs from the energies of the quasistationary states by an amount greater than the width of these states, then the transitions which are accomplished by the electron have a virtual nature (the transitions can originate both in the continuum and in the spectrum of bound states). Equation (1) is equivalent to the relation which serves as the basis for description of the ionization process by perturbation theory in the first nonvanishing order, in this case k_0. The problem of perturbation theory is the calculation of the dependence of the multiphoton cross section $\alpha_{k_0}(I, \omega)$ on the frequency and degree of polarization of the light, and also on the spectrum of the atom.

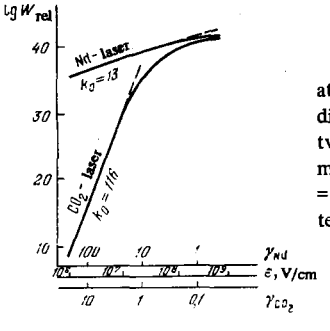

FIG. 1. Ionization of the hydrogen atom in a variable field (calculation according to Keldysh [1]). Data are shown for two typical high-power lasers—neodymium glass, $\lambda = 1.06\ \mu$, and CO_2, $\lambda = 10.6\ \mu$. γ is the adiabaticity parameter (see Eq. 4).

c) The resonance ionization process. By smoothly varying the frequency of the light, we can achieve conditions in which the energy of some number of photons k will equal the energy of any quasistationary state of the electron in the atom, and an intermediate resonance arises. A one-photon resonance with an excited state has been well studied in quantum mechanics (the theory of natural line width, resonance scattering, and resonance fluorescence). However, the intermediate multiphoton resonance arising in ionization has three qualitative differences. First, since the field is always strong, the perturbation of the bound electronic states is important. Therefore not one parameter (frequency), as in a weak field, but also a second parameter (field strength) determine the condition for appearance of resonance. For a fixed frequency a resonance can be induced by the field. Second, the selection rules for multiphoton transitions differ from the well known selection rules for one-photon transitions, being the sum of the selection rules for each absorbed photon. Therefore for an even number of absorbed photons transitions are possible between states with identical parity.[2,15] Third and finally, the very fact of appearance of a resonance ionization process shows that induced transitions from a resonance state to a state with higher energy (including the continuum) are dominant in comparison with spontaneous relaxation. Under these conditions, in spite of the existence of a resonance, the field does not mix the ground state and the resonant state.[27]

Determination of the conditions for realization of the direct and resonance processes is one of the main problems of the investigations. It is apparent that to solve this problem it is necessary to know the spectrum of quasistationary states of the system atom + light field.

d) Spectrum of quasistationary states of the system + light field. In contrast to a constant field, under the action of which, as is well known, the perturbation of the atomic levels reduces to the linear Stark effect (in the case of degenerate states) and to the quadratic Stark effect (in the case of nondegenerate states), the perturbation in a variable field has a significantly more complicated nature. Specifically, the variable field can have various degrees of polarization, the perturbation in a variable field can have both a nonresonance and a resonance nature, and quantum properties (coherence) of the field can appear. The systematic discussion of all phenomena arising in perturbation of an atomic spectrum in a light field is beyond the scope of the present review.[3)] We will discuss briefly below some of the most important cases, information on which is necessary for analysis of experimental data on multiphoton ionization of atoms.

If the initial stationary state s is not degenerate, the perturbing field is not resonant with the energy of any transition $s \to i$ and is linearly or circularly polarized, and the change of energy of this state under the action of the field $\Delta E_s \ll \omega_{si}$, where i is any other state in the unperturbed spectrum, then the quasistationary state of

the system atom + light field is characterized by the same quantum numbers as the initial stationary state, and its energy differs from the energy of the stationary state by a small amount

$$\Delta E_s = -\alpha_s \mathscr{E}^2. \quad (5)$$

In Eq. (5) α_S is the dynamic polarizability of the atom in state s, which is determined by the sum of an infinite number of virtual transitions from the state s to all bound and free states:[4)]

$$\alpha_s = S \frac{(E_i - E_s)\,|V_{is}|^2}{(E_i - E_s)^2 - (\hbar\omega)^2}, \quad (6)$$

where S designates summation over the discrete spectrum and integration over the continuum, and V is the interaction matrix element.[5)] The width of the state s is determined by the total probability of a transition induced by the field from a given state to all remaining states including the continuum. By analogy with the case of a constant field (see Eq. (5)) the perturbation in this case is sometimes called the Stark effect in a variable field. This designation is unfortunate, since in fact the analogy refers only to Eq. (5) and, as can be seen from Eq. (6), the dynamic polarizability differs qualitatively from the polarizability, being a function both of frequency and polarization of the radiation. Therefore, in particular, the main property of the polarizability–an increase with increase of the principal quantum number of the state–cannot be satisfied in the case of the dynamic polarizability. Considerable attention has always been attracted by just this case of perturbation in a variable field, since it is most frequently achieved experimentally (a weak, nonresonant field!), and its theoretical analysis can be carried out by the methods of perturbation theory. Up to the present time, calculations of the dynamic polarizability have been carried out for a large number of atoms in various states.[3] The number of experiments in which a perturbation quadratic in the field has been observed is small[4] (see also Chap. 5). Therefore, although the results of these experiments are satisfactorily described by calculations made by the methods of perturbation theory, it is nevertheless premature to draw final conclusions on the accuracy and most of all on the limits of applicability of these calculations. Equation (5) is the first term of the expansion of ΔE in the intensity of the radiation. With increase of the field strength, higher terms can contribute, determining the so-called hyperpolarizability of the atoms.[5] The equality of the terms $\mathscr{E}^2$ and $\mathscr{E}^4$ can indicate only the inapplicability of perturbation theory for such high field intensity. Studies of hyperpolarizability are at present in their initial stage, so that it is of undoubted interest to obtain any results, either theoretical or experimental.

In all remaining cases the perturbation of the bound states in a variable field has a qualitatively different nature. Thus, under the action of a variable field, quasistationary states can be formed as the result of mixing of the initial stationary states. Here, just as the energy of the states formed can differ substantially from the energy of the initial states, the quantum numbers can also change.

It is well known that a resonance field can be the cause of mixing of the initial stationary states. If the condition $E_S - E_i = \hbar\omega$ is satisfied (where ω is the frequency of the perturbing field), then the quantized system transfers from one state to the other with a frequency Ω_1 proportional to the magnitude of the perturbation. For a sufficiently small width of the resonating states s and i, and sufficiently high field strength, where $\Omega_1 \gtrsim \gamma_{si}$, the probabilities of finding the system in these states become of the same order of magnitude, and a saturation effect arises. A saturation effect can arise also in a multiphoton resonance between the states s and i. However, the condition for appearance of saturation is always internally inconsistent–an increase of the external field strength leads both to an increase of the transition freqeuncy Ω and to an increase in the width of the resonance states. From the relation for a multiphoton resonance $\Omega_k \sim \omega(\mathscr{E}/\mathscr{E}_{at})^k \gtrsim \gamma_{s,i}(\mathscr{E})$, which was obtained in ref. 6, it is easy to estimate that mixing can arise only for a small degree of nonlinearity k. The occurence of resonance mixing also inhibits the process of ionization from the resonating states. If the ionization probability $W_{s,i}(E) \gtrsim \Omega_k$, then it is evident that no mixing can occur.[27]

Mixing of stationary states can occur also in a nonresonance field when the level shift reaches a value comparable with the distance between levels, $\Delta E_S \sim \hbar\omega_{si}$.[7] In this case the possibility of mixing is determined by the duration of the perturbation τ. If $\Delta E_S \tau \ll \hbar$, then the electron at a time t cannot succeed in transferring from a state s to the neighboring state i, and mixing does not occur. It is evident that nonresonance mixing plays an important role in the case of degeneracy of the initial stationary states, i.e., in the hydrogen atom and in the case of sufficiently high hydrogenlike states.[8a,9] In the secular equation, which describes in this case the spectrum of quasistationary states, mixing of the initial states and nonconservation of orbital angular momentum are due to the nondiagonal terms. For frequencies of the order of and smaller than atomic frequencies, the nondiagonal terms have the same order of magnitude as the diagonal terms[9] (the latter are dominant only in the asymptotic limit as $\omega \to \infty$). Therefore, in particular, the perturbation reduces to a linear dependence on field strength (the linear Stark effect) only for a definite frequency of the variable field, for example, at radiofrequencies; in the optical region for most bound states (except the very lowest) the effect is quadratic in nature.

In partially polarized fields a mixing arises of stationary states having different magnetic quantum numbers.[10]

Finally, we note that, depending on the degree of polarization of the light field, different quantum numbers turn out to be good. Thus, for example, for circular polarization the only unique distinguished direction is the propagation vector, and we can speak of the projection of the angular momentum only onto this direction (in contrast to the projection of the angular momentum onto the direction of the field in the case of linear polarization).

Other features of the spectrum of quasistationary states are indicated by consideration of the problem of perturbation of the atomic spectrum by the quasienergy method.[6)] The general solution of the Schrödinger equation with a Hamiltonian periodically dependent on time[8] shows that in a sufficiently strong field the probability of finding the system in the states with energy $E_S \pm k\hbar\omega$ is comparable with the probability of finding it in the state E_S. For a nonresonance light field, estimates[8a] give for k = 1 a necessary field strength $\sim 10^8$ V/cm. In the presence of a quasi-resonance, naturally, a lower

field strength is necessary; a calculation[11] gives a value $\sim 10^7$ V/cm. These estimates show that the appearance of harmonics can play a role under conditions completely achievable experimentally. It is important to note that since the energy of the harmonics $E_S \pm kh\omega$ can be both greater and less than the initial-state energy E_S, on appearance of harmonics of different initial states E_S, E_i, E_j the very concept of greater or lesser energy is lost. The question of the role of quasienergetic states in fields of optical frequency is presently the subject of discussion. For an answer to this question it is necessary to take into account the damping of these states, which is determined, as always in a strong field, by induced transitions, including transitions to the continuum. This problem is still unsolved. On the one hand, use of perturbation theory is completely unjustified not only because of the high field strength, but first of all because of the strong perturbation of the bound electron states, whose energy and also other quantum numbers do not correspond to the data known for the unperturbed spectrum. On the other hand, in most cases (with the exception of those states whose energy is very close to the boundary of the continuum), the transition to the continuum does not have the nature of tunneling, and the adiabaticity parameter has an intermediate value $\gamma \sim 1$. It is necessary to solve the problem of one-photon ionization from excited states exactly, outside the framework of perturbation theory.

If we are to sum up as a whole the situation which exists at the present moment in investigation of the perturbation of an atomic spectrum by a light field, we can state that there is as yet no systematic theoretical description of the whole diversity of phenomena arising. No better situation exists with the experimental data, which have been obtained so far only for individual cases–the quadratic Stark effect and single-photon resonance in a two-level system.

e) Processes leading to ionization of an atom in a strong light field. A brief discussion of the specific features of processes associated with absorption of many photons by an atomic electron permits qualitative classification of the various processes leading to ionization of the atom.

Direct multiphoton ionization occurs under conditions in which the parameter $\gamma \gg 1$ and intermediate resonances do not arise. The probability of the direct process is associated with the field strength by the power relation (1).

Resonance ionization occurs when an intermediate resonance appears between the energy $1 < k < k_0$ of the photons and the energy of a bound electron state in the atom. On appearance of an intermediate resonance the ionization process is determined substantially by perturbation of the resonance state in the light field.

Tunneling ionization occurs for a parameter value $\gamma \ll 1$; it is described by an exponential relation between the ionization probability and the field strength of the form (3) and is characterized by the absence of a dependence of the ionization probability on the frequency of the radiation.

2. THEORETICAL METHODS OF DESCRIBING THE IONIZATION OF AN ATOM IN A LIGHT FIELD

a) Methods used and their regions of application. The specific features of the ionization process discussed in the preceding chapter indicate those methods which can be used to describe the ionization process in a strong light field. When using different methods, as always, it is necessary to analyze the conditions of their applicability for solution of each specific problem. The large number of parameters which determine the process of ionization of atoms by light, as a rule, hinders the definition of well understood criteria.

Let us turn first to perturbation theory. A single criterion of applicability of perturbation theory for description of multiphoton processes can hardly be formulated, if only because of the complex nature of the perturbation of the atomic spectrum by the light field. However, it is possible to make some obvious evaluations.[7] We will consider the standard approach of perturbation theory in which the unperturbed atomic spectrum is taken as the basis, and in the final state it is assumed that the electron is in the Coulomb field of the residual atom. With this formulation of the problem for the final state–a free electron removed from the atom–the condition $\Delta p/p \ll 1$, where p is the momentum of the emitted electron whose energy $E_{kin} \leq \hbar\omega$, and Δp is determined by the oscillation energy E_{osc} in the field of the wave, gives an upper limit of the light field intensity in the form[12]

$$\mathscr{E} \ll \mathscr{E}_{at}/k_0^{3/2}. \tag{7}$$

(Inclusion of the Coulomb field of the residual atom should evidently somewhat weaken this criterion.) Numerical calculations carried out in ref. 9 showed that nonresonance perturbation of bound states in the hydrogen atom already for $n \geq 4$ arrives at the asymptotic value corresponding to perturbation of a free electron. Thus, criterion (7) can be used also for the case of perturbation of bound states (except the very lowest). If we assume that a typical degree of nonlinearity in ionization of atoms by light has a value $k_0 \sim 10$, then it follows from Eq. (7) that perturbation theory is applicable for a field strength much less than 10^8 V/cm. Since the ionization of atoms in a strong light field has a multiphoton nature ($\gamma \gg 1$) up to field strengths of the order 10^8 V/cm (for the same value $k_0 \sim 10$), there is a definite interval of field strength 10^7–10^8 V/cm in which, although the process has a multiphoton nature ($\gamma \gg 1$), standard perturbation theory is probably not applicable for its description.

The quasiclassical method is widely applied to description of the ionization of an atom in a strong light field. Actually, the strong field corresponds to a high intensity of the photon flux. In the case of frequencies in the optical region, we have $\omega \ll \omega_{at} = I/\hbar$. If the field strength $\mathscr{E} \ll \mathscr{E}_{at} = 5 \times 10^9$ V/cm, then the ionization occurs slowly in comparison with atomic times, and the barrier is wide. Finally, since the wavelength of light is much greater than the size of the atom, the field can be considered uniform over the extent of the atom. Comparison of the criteria of applicability of the quasiclassical approximation

$$\mathscr{E}/\mathscr{E}_{at} \ll 1, \ \omega/\omega_{at} \ll 1 \tag{8}$$

with the criterion of applicability of perturbation theory (7) shows that use of the quasiclassical approximation can in principle permit an advance into the region of high field strength.

The quasienergy method in formulation of the problem itself does not contain limitations regarding strong fields–the Schrödinger equation with a Hamiltonian which is a periodic function of time is solved. However,

the classical approach used in the quasienergy method–solution of the problem into a spectrum with subsequent account of damping–can hardly be considered justified in the case in which the field is strong; the probabilities of transitions in a strong field are always high, so that the damping cannot be assumed small.

It is possible to identify a number of other methods, but they do not have a general nature. The absence of generality is not in itself a deficiency important in practice. After the work of Keldysh[1] it became clear that, depending on the specification of several basic parameters characterizing the atom and the field, the ionization process has a qualitatively different nature. Basic practical interest is therefore presented not only by the problem of creating a sufficiently general theoretical method of describing the ionization process, but also by obtaining particular solutions, by clear determination of the conditions of their applicability, and by accurate account of the approximations made.

Let us now discuss some specific results obtained by various methods of calculation.

b) The perturbation theory method. The absorption by an atomic electron of k_0 photons can be described in the first nonvanishing (k_0) approximation of nonstationary perturbation theory in the case in which the electron transitions are virtual. In terms of nonstationary perturbation theory of order k_0, the probability of a k_0-photon process of ionization of the atom is described by the following relation[8]:

$$dW = \frac{m}{\hbar (2\pi)^2} (2\pi\alpha F\omega)^{k_0} | K_{0f}^{(k_0)} |^2 d\Omega_k, \qquad (9)$$

where $K_{0f}^{(k_0)}$ is the compound matrix element of the electron transition from the ground state to the continuum, which contains a $(k_0 - 1)$-fold summation over the entire set of bound and free states and has the form

$$K_{0f}^{(k_0)} = \sum_{n_1} \sum_{n_{k_0-1}} \frac{V_{0n_1} \dots V_{n_{k-1}n_k} \dots V_{n_{k_0-1}f}}{[E_{n_{k_0-1}} - E_0 - (k_0-1)\hbar\omega] \dots [E_{n_k} - E_0 - k\hbar\omega] \dots [E_{n_1} - E_0 - \hbar\omega]}. \qquad (10)$$

In Eq. (10) $V_{n_{k-1}n_k}$ is the dipole matrix element which in the nonrelativistic approximation describes the transition between the states n_{k-1} and n_k having energies $E_{n_{k-1}}$ and E_{n_k}; E_{n_k} are the energies of the stationary bound states of the electron in the absence of the field; in the final state the action of the light field on the electron is neglected. Equation (9) does not describe the ionization process for those radiation frequencies corresponding to occurrence of resonances; the energy denominators for the resonance frequencies in Eq. (10) go to zero, and $K_{0f}^{(k_0)} \to \infty$; in the first nonvanishing approximation, damping can be taken into account only phenomenologically.

Putting off for some time discussion of the methods of calculating the infinite sum (10), let us first analyze qualitatively the typical result of such a calculation, shown in Fig. 2–the dependence of the ionization probability on the frequency of the radiation. The data refer to 3-photon ionization of the hydrogen atom. For the hydrogen atom the spectrum of s states is identical to the spectrum of d states, and therefore the resonances for circularly and linearly polarized radiation coincide except for the resonance at n = 2, which occurs only for linear polarization. The general nature of the dispersion differs sharply from the similar dependences for one-photon processes–the width of the maxima is many orders greater than the width of the resonance states and may reach values of the order of the distance between resonances. This nature of the dispersion function is typical for multiphoton processes, since the compound matrix element (10) is determined by the sum of many matrix elements for individual virtual transitions. We should note the qualitative difference of the dispersion functions for light of different polarizations. In the case of circular polarization, in each region between resonances there is a frequency at which the ionization probability is zero. These minima reflect the specific behavior of circularly polarized light–in absorption of each photon the orbital angular momentum of the electron constantly increases. Thus, there is no systematic preference for a definite polarization; depending on frequency, the ionization probability for a given polarization may be both greater and smaller than for another polarization.[9]

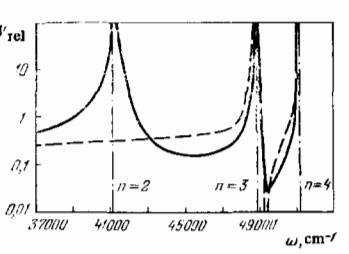

FIG. 2. Hydrogen atom ionization probability W_{rel} (in relative units) as a function of light frequency (ω calculation by standard perturbation theory [21]). Solid line–linearly polarized light, dashed line–circularly polarized light; n is the principal quantum number of the bound electron state.

We will now turn to methods of calculating the transition probability. In calculation of the compound matrix element (10), two difficulties arise.

The first difficulty is traditional and consists in the necessity of constructing the wave function of the optical electron in a complex atom. (This difficulty, naturally, is absent in solution of the problem of ionization of the hydrogen atom.) By analogy with one-photon processes, one uses in the corresponding modification the quantum defect method[13], the Hartree-Fock method, and the pseudopotential method. The quantum defect method, as is well known, is based on the fact that in the matrix elements the main contribution is from regions far from the nucleus, where the effective single-particle potential can be considered Coulomb with the charge of the residual ion. We should therefore expect that use of the quantum defect method should give gest results in the case of ionization of the alkali atoms. Obtaining an exact solution for a pseudopotential in analytic form is impossible. Therefore, use of the pseudopotential method reduces essentially to a phenomenological choice of analytic potentials, with a definite accuracy of the modeling pseudopotentials.[14]

The second difficulty is specific for multiphoton processes; it is based on the virtual nature of the transitions occurring and consists in the necessity of infinite summation in calculation of the compound matrix element (10).

The summation methods used can be divided into phenomenological methods (the methods of Gold, Bebb, and Morton) and exact methods (the Schwartz-Tiemann method and the Green's function method).

Gold's method[15,16] is based on the empirical assumption of the possibility of introducing an average energy instead of the infinite number of energies $E_0 - E_{n_k}$ (see Eq. (10)). Numerical calculation has shown that the energy of the first excited state $E_{2p} = 10.2$ eV is a satis-

factory value of the average energy in the hydrogen atom. By analogy, the energy of the first excited state was taken as the average energy also in the case of complex atoms. Thus, calculations have been made of the ionization probability of the hydrogen atom up to $k_0 = 7$, and for noble gas atoms[15] up to $k_0 = 14$. Bebb's method is based on summation of a finite number of terms providing the greatest contribution to the compound matrix element. Inthis way theprobabilities of 2–3 photon processes for ionization of alkali atoms[16] have been calculated. Morton's method[17] is based on the directly opposite but equally empirical assumption of the possibility of averaging all matrix elements, which in order of magnitude are assumed equal to

$$V_{n_{k-1}n_k} = \sqrt{\frac{\hbar^2}{2mI}}\sqrt{\frac{I}{E_{n_{k-1}n_k}}},$$

where the first factor is the size of the atom. These methods have been used to calculate the probabilities of multiphoton ionization of the hydrogen atom, the alkali atoms, mercury, and the noble gas atoms by radiation of ruby and neodymium lasers and the corresponding second harmonics.[17] An obvious deficiency of any of the phenomenological methods is the unknown accuracy of the calculations.

A number of calculations of ionization of the hydrogen atom have utilized the Schwartz-Teimann method[18], which permits reducing the problem of the infinite summation to integration of a chain of coupled inhomogeneous differential equations of first order. Although this method is not empirical and is mathematically well justified, the calculations turn out to be complicated, which can be seen in the example of calculation of the two-photon ionization of the hydrogen atom.[19]

Calculations based on use of the Green's function for description of the optical electron[10] appear the most promising. The Green's function $G_E(\mathbf{r}, \mathbf{r}')$ of the Schrödinger equation is defined by the relation

$$G_E(\mathbf{r}, \mathbf{r}') = \sum \frac{\Psi_{n_k}(\mathbf{r})\Psi^*_{n_k}(\mathbf{r}')}{E_{n_k} - E} + \int \frac{\Psi_{E'}(\mathbf{r})\Psi^*_{E'}(\mathbf{r}')}{E' - E}\,dE',$$

where $\Psi_{n_k}(\mathbf{r})$ and $\Psi_E(\mathbf{r})$ are the wave functions of the optical electron in the discrete and continuous spectra. The compound matrix element (10) written with use of the Green's function has the form:

$$K_{0f}^{(k_0)} = \langle f | (e, \mathbf{r}_{k_0}) G_{E_i+(k_0-1)\omega}(\mathbf{r}_{k_0}, \mathbf{r}_{k_0-1}) (e, \mathbf{r}_{k_0-1}) \ldots G_{E_i+\omega}(\mathbf{r}_2, \mathbf{r}_1)(e, \mathbf{r}_1) | i \rangle. \quad (10')$$

Since $G_E(\mathbf{r}, \mathbf{r}')$ satisfies the Schrödinger equation, then by letting the operator $\hat{H}$ act on the Green's function, we obtain the following linear inhomogeneous equation:

$$\hat{H}G_E(\mathbf{r}, \mathbf{r}') = -\delta(\mathbf{r} - \mathbf{r}').$$

Thus, if we introduce the Green's function, calculation of the probability reduces to solution of this equation, in integration of which, in particular, it is possible to use various approximate methods developed in atomic quantum mechanics. This method permits in principle the exact summation of the infinite sum (10). Here the spectrum of the atom (in the absence of a field) is taken into account exactly, since the poles of the Green's function coincide with the bound electron states. For the optical electron in a complex atom the Green's function was constructed in the approximation of the quantum defect method.[13] Use of the Green's function method permitted calculation of the probability of the direct multiphoton ionization to be carried out over a wide range of frequency of the radiation for the hydrogen atom in the interval[20] $2 \leq k_0 \leq 16$, and for the alkali atoms in the interval[21] $2 \leq k_0 \leq 5$.

Most of the calculations were made for a linearly polarized field. The question of the dependence of the ionization probability on the degree of ellipticity of the radiation has been discussed by several authors,[21,22] who obtained both numerical data for circular polarization and the dependence of the ionization probability on the degree of ellipticity of the light.

The enumeration of the various methods of calculation of the cross sections for the direct process of multiphoton ionization, carried out in terms of perturbation theory, raises the question of how optimal these methods are. The nonanalytic nature of the calculations, which require use of computers, and also the use of various approximations, makes it difficult to obtain an answer to this question. However, comparison of the results of the different calculations, carried out for the same cases, permits some general conclusions to be drawn. In Tables I–IV we have given the values of the multiphoton cross sections calculated by various methods with various simplifying assumptions.[11)]

The example shown in Table I of calculation of the cross section for six-photon ionization of the hydrogen atom illustrates the use of different methods of summation–the cross sections differ by an order of magnitude.

No less a difference is obtained in calculation of the cross sections for ionization of complex atoms (see Table II) (note that in this case we have listed α_3, and in the case of ionization of hydrogen–α_8).

The sensitivity of the calculations to the choice of wave functions is illustrated by the data given in Table III.

In addition to internal consistency and the possibility of systematic monitoring of the various steps of the calculation, an obvious criterion of optimality of a given method is the result of comparison with experimental data. Such a comparison has been made in Chap. 4, where we also discuss the question of the limits of applicability of perturbation theory in the field strength.

Calculation of the probability of multiphoton excitation–of a bound-bound virtual multiphoton transition in a strong light field–is complicated, in comparison with calculation of the ionization process, by the necessity of taking into account the change in the transition energy, and also the width of the final state under the influence of the radiation field. Dynamic effects associated with the perturbation of the resonance state s play a role in the case when $\Delta E_s(\mathscr{E}) \geq \Gamma_k, \gamma_s(\mathscr{E})$, where Γ_k and $\gamma_s(\mathscr{E})$ are the reduced width of the radiation line and the width of the resonance state s (it is assumed that the perturbation of the resonance state is greater than that of the ground state (see Chap. 5)). The very fact of taking into account the line width of the radiation leads to the necessity of solving the problem of the transition of the electron from a state o to the state s as the result of absorption of k_0 photons with various frequencies. Since the value of the compound matrix element remains practically constant as the frequency varies within the limits $\Gamma_k, \gamma_s(\mathscr{E})$, the calculation is separated into two steps–calculation of the compound matrix element and inclusion of the perturbation of the resonance state by the radiation field. The expression for the probability depends on the shape of the radiation line and the resonance state. The compound matrix elements of bound-bound multi-

TABLE I. Theoretically calculated cross sections for six-photon ionization of the hydrogen atom α_6 for $\hbar\omega = 2.36$ eV, carried out by various methods (linear polarization of the light $[\alpha_6] = cm^{12} sec^5$)

$\lg \alpha_6$	Method of calculation	Reference
−179.7	Green's function method	20a
−180.1	Gold's method	15
−181.4	Morton's method	17

TABLE II. Theoretically calculated cross sections for three-photon ionization of the cesium atom α_3 for $\hbar\omega = 1.78$ eV, carried out by various methods (linear polarization of the light, $[\alpha_3] = cm^6 sec^2$)

$\lg \alpha_3$	Method of calculation	Reference
−75.0	Green's function method in quantum defect approximation	21
−76.0	Bebb's method	16
−76.0	Morton's method	17

TABLE III. Theoretical calculations [21] of cross sections for direct multiphoton ionization, carried out with ground-state wave functions constructed by the quantum defect method (QDM) and the Hartree-Fock method (H-F) (values of $\log\alpha_{k_0}$ are given; $[\alpha_{k_0}] = cm^2 k_0 sec^{k_0-1}$)

k_0	4	4	5
Atom	K	Rb	Na
QDM	−106.4	−106.9	−139.4
H-F	−107.8	−107.0	−137.4

TABLE IV. Compound matrix elements $K_{is}^{(k)}$ for multiphoton bound-bound transitions (theory of ref. 23)

k	2	2	3	3	4
Atom	He*	Cs	K	Cs	Na
$\hbar\omega$, eV	1.78	1.78	1.17	1.17	1.17
Transition	2^1s-6^1s	$6s-9d$	$4s-4f$	$6s-6f$	$3s-7s$
$K_{is}^{(k)}$, Atomic units	$4.7\cdot10^3$	$1.0\cdot10^5$	$1.8\cdot10^{12}$	$1.5\cdot10^{11}$	$7.4\cdot10^{15}$

photon transitions calculated by the Green's function method are listed in Table IV.

The difference in the value of the matrix elements for three-photon excitation of potassium and cesium atoms demonstrates the role of the real spectrum of the atom and the impossibility of using a hydrogen-like model.

Remaining within the framework of the standard perturbation theory approach, it is also possible to carry out a calculation of the ionization probability in the presence of an intermediate resonance, if we utilize a model in which the transition from the ground state to the continuum can be represented in the form of two successive transitions, and the nature of the perturbation of the resonance state is known. The ionization probability in this case is equal to the product of the transition probabilities, each of which is calculated independently (see refs. 1, 24a, 24b, and then Chap. 5).

However, we will not adopt the standard perturbation theory approach in principle for description of dynamic effects in the presence of an intermediate resonance–a dependence of the ionization probability on the frequency and intensity of the light field–since the nature and magnitude of the perturbation of the resonance state change as these parameters change. In the compound matrix element (10) the corresponding denominator $(E_S - E_0 - k\hbar\omega)$ in the presence of a resonance goes over to $(E_S(\mathscr{E}) - E_0 - k\hbar\omega - i\gamma_S(\mathscr{E}))$. Since with variation of $\mathscr{E}$ the dependence of the probability on $\mathscr{E}$ changes, the process can be described only in a higher order of perturbation theory (higher than k_0). Description of the ionization process in the presence of a resonance can be accomplished by using as a basis the spectrum of the system atom + field. Here the main problem lies in constructing the Green's function for this system. Use of this method is well known both for description of one-photon transitions[25] and for multiphoton transitions[26]. Solutions have been obtained only in a number of particular cases. Although in principle the promise of this method is obvious, it is premature at the present to make any conclusion of its practical value.

In concluding our brief review of the results of application of perturbation theory methods for description of multiphoton ionization of atoms, it is necessary to formulate the main conclusions. Up to the present time there has been an extensive development of the methods of standard perturbation theory of k_0-th order, in which the unperturbed spectrum of the atom is taken as a basis, and the action of the field on the electron in the final state is neglected. Standard perturbation theory can describe only the direct process (ionization or excitation). In principle in an arbitrarily weak variable field, conditions of perturbation of the bound electron states can be realized such that the electron transition cannot be described in terms of the standard perturbation theory approach.

c) The quasiclassical method[12]. A number of workers[1,28,30] have solved the problem of ionization in a variable field under the conditions where the quasiclassical criteria (8) are satisfied. The most systematic use of the quasiclassical method is represented by the calculation of the quasiclassical trajectory of a subbarrier transition of an electron[28] and the quasiclassical wave function[30]. It is evident that in the adiabatic case ($\gamma \ll 1$), where the ionization occurs in times small in comparison with the period of variation of the field, the ionization probability can be calculated by averaging over the period of the ionization probability in a constant crossed field[27] ($|\mathscr{E}| = |\mathbf{H}|$, $\mathscr{E} \perp \mathbf{H}$).

Use of the quasiclassical method has permitted a general solution to be obtained only in one case–for removal of an electron from a short-range potential well. The probability of this process is

$$W \approx \left|\frac{\mathscr{E}}{\mathscr{E}_{at}}\right|^2 \exp\left[-\frac{2\omega_{at}}{\omega} f(\gamma)\right], \quad f(\gamma) = \left(1+\frac{1}{2\gamma^2}\right)\operatorname{Arcsh}\gamma - \frac{\sqrt{1+\gamma^2}}{2\gamma}. \tag{11}$$

The function $f(\gamma)$ increases monotonically with increasing field strength, and therefore for a fixed radiation frequency the ionization probability increases with increasing field. For $\gamma \ll 1$, we have $f(\gamma) \sim \gamma$, and Eq. (11) with an accuracy to the preexponential factor goes over to the well known formula (3) for a constant field $W \sim \exp(-C/\mathscr{E})$. For $\gamma \gg 1$, we have $f(\gamma) \sim \ln 2\gamma - 1/2$, $W \sim (1/2\gamma)^{2\omega_{at}/\omega} \sim F^{k_0}$, which is equivalent to Eq. (1).

A difficulty which it has not yet been possible to overcome is to take into account the Coulomb field of the residual atom. Because of the long range nature of the Coulomb field, it distorts the electron motion at distances large in comparison with the atomic radius. The essence of the difficulty arising is obvious–it is necessary to take into account the action of two fields on the electron in the final state. The action of the Coulomb field has so far been taken into account only by the perturbation theory method; here it is assumed that the action of the Coulomb field is a small correction to the action of the field of the light wave.[28c] This naturally limits the region of applicability of the results obtained in the direction of weak light fields, since it is necessary that the following condition be satisfied:

$$\gamma \ll \sqrt{\frac{\mathscr{E}_{at}}{\mathscr{E}}}. \qquad (12)$$

Even for the maximum field strength at which the direct multiphoton ionization process has been observed[42], $\sim 10^8$ V/cm, we have $\gamma \sim 5$, while Eq. (12) reduces numerically to $\gamma \ll 10$. Thus, strictly speaking, a general quasiclassical description of the multiphoton ionization of a real atom cannot be applicable for existing experimental data. It is necessary to have in mind that, as the calculations of ref. 28c have shown, with satisfaction of the criterion (12) the inclusion of the Coulomb field increases the ionization probability by a factor of the order $(2\mathscr{E}_{at}/\mathscr{E})^2$. In ref. 28a the authors succeeded in taking into account the Coulomb field without limitations on the light field strength, but only for circular polarization of the radiation. This may reflect the fact that in absorption of a large number of photons of the circularly polarized field, the angular momentum of the electron is always large, the electron is far from the nucleus, and the effect of the Coulomb field is small in comparison with the case of linear polarization. Interest is evident in advancing the quasiclassical method to the region of lower intensities of the linear field and higher values of the parameter γ. Here, however, we must have in mind that by the very essence of the quasiclassical approach to solution of the ionization problem, this method, which originates from averaged characteristics, cannot describe the appearance of intermediate resonances. Therefore we should not expect success in application of the quasiclassical method for $\gamma > 1$ in those cases where k_0 is not very large or in which the bound states are located not too high, and therefore it will not very often be successful.

The quasiclassical method has also been applied to description of the multiphoton excitation of the atom. Since, for multiphoton transitions, the external field can be considered as an adiabatic perturbation and the wave function is quasiclassical in the time, then, as is well known[13], the problem of finding the wave function at the moment $t \to \infty$ from its known value at the moment $t = 0$ is equivalent to the problem of reflection above the barrier. Using this analogy, Zaretskiĭ and Kraĭnov[30] obtained an expression for the probability of multiphoton excitation in a two-level system for $\hbar\omega \ll E_i - E_j$, having the form

$$W = (A\mathscr{E})^{2k_0} \exp(Bk_0\mathscr{E}^2), \qquad (13)$$

where A and B are constants which do not depend on the field strength. In Eq. (13) the first factor corresponds to the use of perturbation theory, and the second to the correction which the quasiclassical method makes it possible to obtain. In application of this method to calculation of bound-bound transitions in a real atom, it is necessary to determine the adiabatic energies of the initial and final states as a function of time.

d) Conclusion. We will briefly review the possibilities existing presently for theoretical description of the various processes leading to ionization of atoms in a strong light field.

The direct ionization process at a field strength $<10^7$ V/cm–perturbation theory is applicable; many calculations have been made for various specific cases.

The direct ionization process for field strengths $>10^7$ V/cm–there is no general solution of the ionization problem; particular solutions have been obtained with additional limiting assumptions.

Tunneling (field strength $>10^8$ V/cm)–a general solution of the problem has been obtained by the quasiclassical method.

The resonance ionization process–results for individual particular cases have been obtained qualitatively and quantitatively.

It must be noted that the successes in the theory are for those cases in which the spectrum of bound electron states does not play an important role (tunneling, the direct process). On the other hand, those cases in which the main effect reduces to perturbation of the bound states are a natural difficulty for the theoretical description, particularly in the case of strong perturbation. There is obvious interest in the development of methods going beyond standard perturbation theory. Individual advances have been made in this direction–the quasienergy method [8a,8b,11], construction of the Green's function for the system atom + light field[25,26], and the solution for a quantum system having a constant dipole moment[29]. However, the conditions of applicability of these particular results have not been sufficiently justified and the validity of the approximations made has not been carefully analyzed. The latter remark refers also to the results obtained in the quasiclassical approximation[1,28], and to the account of perturbation of the spectrum of bound states. Finally, it should be noted that recently[30,31] there has been a rigorous demonstration of the internal inconsistency of the gauge transformation method (momentum-translation approximation[32]), which has been extensively used for solution of many problems without the necessary attention to its justification.

3. FORMULATION OF EXPERIMENTS

An experiment to study multiphoton ionization consists of irradiating a target of neutral atoms by a laser beam and detecting the ions produced. Here it is possible to vary the parameters characterizing the radiation field (intensity, frequency, spectrum width, polarization, coherence), and also the atom studied.

a) Competing effects. There are fundamental limitations associated with the possibility of appearance of competing effects which also lead to formation of ions. In a strong light field such an effect is ionization by accelerated electrons. A free electron can acquire the energy necessary for ionization by means of the inverse bremsstrahlung effect or the induced Compton effect. In the case of inverse bremsstrahlung, the electron must undergo a series of successive elastic collisions with neutral atoms, and therefore the acquisition of energy

from the field is determined by three parameters–the field strength, the length of the period during which the field acts on the electron, and the density of neutral atoms. Analysis of the experimental and theoretical data permits us to write down the following approximate condition which the electron must satisfy in order to acquire sufficient energy for ionization: $n\tau\mathscr{E}^2 > 10^{23}$ (where the density of neutral atoms n is expressed in cm^{-3}, the duration of field action on the electron τ in sec, and the electric field strength in V/cm). In the case of the induced Compton effect, the reradiation of the photon absorbed by the electron occurs not spontaneously but under the influence of the external field, the probability is proportional to the square of the radiation intensity, and the reradiated photon has a frequency and direction of propagation determined by the field. Since the wave is always not ideally monochromatic and not plane, the electron can absorb a photon of another frequency and having another wave vector than those of the inducing field, and the electron energy can increase. Analysis of the available experimental and theoretical data permits us to write the following approximate condition for acquisition by the electron of the energy necessary for ionization: $\tau\mathscr{E}^4 > 10^{24}$ (where the designations and dimensions are the same as above). Estimates with these relations show that for a laser pulse duration $\tau \lesssim 10^{-8}$ sec (a value typical for a solid-state laser with Q switching) it is necessary to limit ourselves to a target density $n < 10^{16}$ (i.e., to use either a rarified gas or an atomic beam), so that ionization by accelerated electrons will not occur. Under these conditions the only mechanism for ion production is ionization by the radiation field.

b) Background effects and their elimination. In the first experiments it was established that it is necessary to detect ions, and not electrons, since the molecules of the residual gas in the vacuum chamber are also easily ionized. The ionization of residual gas molecules has been studied in detail by Chin.[33] It turned out that the main source of ions is complex hydrocarbon molecules. The degree of nonlinearity of the production of these ions lies in the range 2–6 (depending on the frequency of radiation used). Thus, the yield of the ions investigated substantially exceeds the yield of background ions and the background can be neglected only in the case of investigation of relatively few-photon processes. In all remaining cases it is necessary to separate the detected ions carefully from the background, which is accomplished on the basis of the time of flight to the detector.

c) Range of variation of field intensity at which it is possible to study the nonlinear effect. The value of electric field strength at which it is possible to study multiphoton ionization of a certain atom for a fixed radiation frequency is limited on both the low and high frequency sides. The high frequency limitation is due to achievement of 100% degree of ionization in the interaction volume, i.e., satisfaction of the condition

$$\int_0^\tau W(t)\,dt \sim 1 \qquad (14)$$

(where W is the ionization probability per unit time and τ is the duration of the radiation pulse). On the low frequency side the field strength is limited by the condition in which a threshold number of ions (in principle, one ion) is formed in the interaction volume. Accordingly, the interaction volume is important for the lower limit, since the density of neutral atoms is limited on the high side as the result of the possibility of appearance of competing effects. The present state of laser technology permits the necessary field strengths to be accomplished without focusing of the radiation, in volumes of $\sim 1\ cm^3$. Under optimal conditions (volume $\sim 1\ cm^3$, density $\sim 10^{16}\ cm^{-3}$) it is possible to have a range of variation of the number of detected photons from 1 to 10^{16}. Even this range is not at all large for a strongly nonlinear process. For example (see Eq. (1)), the fifteen-photon process under these conditions can be observed at a field strength varied by a factor ~ 3. All experiments carried out so far have been performed under conditions in which the laser radiation was focused by short focal length lenses and the volume in which the necessary field strength was achieved was in the range from 10^{-5} to $10^{-6}\ cm^3$, so that the dynamic range was several times smaller than the optimum value given above. A substantial variation of the field strength at which it is possible to investigate the nonlinear process can be achieved only by varying the degree of nonlinearity of the process, i.e., varying the frequency of the radiation or the ionization potential of the atom studied.

d) Measurement of the degree of nonlinearity, probability, and cross section of multiphoton ionization of atoms.[34a,34b,36] In studying multiphoton ionization the nonlinear nature of the elementary event of absorption of several photons by the atomic electron appears in a substantial way. In this case it is insufficient to measure the classical combination of parameters which must be known to determine the cross section of any effect arising as the result of absorption of one particle (photon): the number (density) of incident particles, the density (number of particles) of the target, and the number of particles produced. In the case of a nonlinear process it is necessary also to measure the intensity of the radiation at each point of the target at each moment of time.

Since the ionization probability is a nonlinear function of the radiation intensity, since the radiation is always distributed nonuniformly over the target, and since the total yield of ions from the entire target is detected, then for an infinitely small degree of ionization the number of ions produced N_i is related to the intensity of radiation by the expression

$$N_i = n\int\int W(F)\,dv\,dt, \qquad (15)$$

where the density of neutral atoms in the target n is assumed uniform and practically unchanged as the result of production of ions. The integration must be carried out over the irradiated target volume and over the duration of the laser radiation pulse. It can easily be seen from Eq. (15) that only when W is a linear function of the radiation intensity is the integral directly proportional to the number of incident photons (the case of one-photon ionization). For a nonlinear dependence of the ionization probability on radiation intensity, before experimentally determining the cross section or probability of the process it is necessary to know the form of the dependence W(F).

If we assume that the probability is related to the intensity by a power law, i.e., Eq. (1) is satisfied, then the expression for the number of ions produced (15) takes the form

$$N_i = \alpha_{k_0} n\int\int F^{k_0}\,dv\,dt. \qquad (16)$$

The intensity of radiation is related to the quantities measured experimentally by the expression

$$F = F_0 f(x, y, z, t) = \frac{Q}{\hbar\omega} \frac{f(x, y, z, t)}{\iint \Phi_z(x, y, t)\, ds\, dt}, \qquad (17)$$

where F_0 is the maximum intensity realized at some point of the target at some moment of time, and Q is the energy in the laser radiation pulse. The function Φ describes the distribution of the radiation for a fixed coordinate z in a plane passing through the point with maximum intensity.

From Eqs. (16) and (17) it follows that the number of ions produced is related to the energy in the laser pulse as follows,

$$N_i = \frac{\alpha_{k_0} n}{(\hbar\omega)^{k_0}} \frac{\iint f^{k_0}\, dv\, dt}{\left(\iint \Phi\, ds\, dt\right)^{k_0}} Q^{k_0} = CQ^{k_0}. \qquad (18)$$

It is evident from Eq. (18) that if in a series of consecutive pulses of laser radiation only the energy Q passing through the target varies, and the space-time distribution of radiation at the target remains constant, then the functional relation $N_i(Q)$ is similar to the functional relation W(F) characterizing the ionization process. In practice both the function f(x, y, z, t) and the function $\Phi(x, y, t)$ do not remain constant, particularly in the case where the laser radiation has a multimode nature. This is clearly visible from the presence of fluctuations in the magnitude of the ion signal for a constant energy of the radiation in a series of consecutive laser radiation pulses. Experiments have shown that the fluctuations are random in nature and the deviations from the mean are distributed according to a normal law. Under these conditions the function $N_i(Q)$ and consequently also the experimentally measured function $A_i(Q)$, where A_i is the amplitude of the ion signal, are equivalent to the desired function W(F). In accordance with Eq. (1), in the case in which the ionization process has a direct nature, the function $A_i(Q)$ should have a form $A_i \sim Q^{k_0}$, where k_0 is the number of photons whose absorption is necessary for ionization. The degree of nonlinearity can be determined from the experimental data from the relation

$$k = \frac{\partial \lg A_i}{\partial \lg Q} \sim \frac{\partial \lg W}{\partial \lg F}. \qquad (19)$$

In accordance with Eqs. (16) and (17), the cross section for the direct process associated with absorption of k_0 photons has the form

$$\alpha_{k_0} = \frac{N_i (\hbar\omega)^{k_0}}{nQ^{k_0}} \frac{\left(\iint \Phi\, ds\, dt\right)^{k_0}}{\iint [f(x, y, z, t)]^{k_0}\, dv\, dt}.$$

The function f(x, y, z, t) cannot be directly measured. The function $\Phi(x, y, t)$ can be measured by use of a high speed photodetector with a sufficiently high degree of space-time resolution, but this method is complicated in practice. Therefore in all experiments independent measurements have been made of the space and time distributions. The dimensionless functions (f(x, y, z, t) and $\Phi(x, y, t)$ can be represented in the form of factors

$$f(x, y, z, t) = \varphi(x, y, z)\,\psi(t), \qquad \Phi(x, y, t) = \eta(x, y)\,\psi(t), \qquad (20)$$

in the case that the change of intensity with time $\psi(t)$ at each irradiated point of the target follows the same law. If Eq. (20) is satisfied the expression for the cross section has the form

$$\alpha_{k_0} = \frac{N_i}{n} \left(\frac{\hbar\omega}{Q}\right)^{k_0} \frac{S^{k_0} \tau^{k_0}}{V_{k_0} \tau_{k_0}}, \qquad (21)$$

where

$$S = \frac{1}{\eta_{\max}} \int \eta(xy)\, ds, \qquad \tau = \frac{1}{\psi_{\max}} \int \psi(t)\, dt,$$
$$V_{k_0} = \int [\varphi(x, y, z)]^{k_0}\, dv, \qquad \tau_{k_0} = \int [\psi(t)]^{k_0}\, dt. \qquad (22)$$

The quantities S and τ are the normalized cross section of the light flux at the target and the normalized duration of the laser pulse. The quantities V_{k_0} and τ_{k_0} play the role of the target volume and the duration of the interaction which are effective for a process involving absorption of k_0 photons in one event. For a nonuniform distribution of radiation intensity over the target and a high degree of nonlinearity of the process, the effective characteristics are substantially different from the corresponding quantities V_1 and τ_1. The choice of normalization of the quantities S and τ can be arbitrary. However, in the case of highly nonlinear processes the standard normalization for linear processes at half the maximum amplitude (the half-width or width at half-height) is not logically justified, since the main contribution to the integral effect is from the regions where the intensity is maximal. Therefore the normalization is carried out to the maximum value of the amplitude.

It should be noted that the expression for the cross section of the nonlinear process (21) is valid if only two assumptions are satisfied–the power law form of the dependence of probability on radiation intensity, and the possibility of separating the space and time variables in the distribution function of the radiation over the target–and therefore this expression describes a broad class of processes of ionization and excitation of atoms and molecules.

The dimensionality of the cross section is determined by the degree of nonlinearity of the process k_0, $[\alpha k_0] = \text{cm}^{2k_0}\ \text{sec}^{k_0-1}$. Therefore it is meaningless to compare the cross sections for processes with different degrees of nonlinearity. It is necessary to compare the probabilities for a fixed value of the radiation intensity.

The accuracy in measurement of multiphoton cross sections by the absolute method described above is relatively low in comparison with the accuracy of measurement of one-photon cross sections. The principal error is due to the high power of k_0 in which the measured quantities S, τ, φ and ψ appear in Eq. (21).

Recently Delone et al.[34b] proposed another method of measuring the cross section which permits the accuracy to be greatly improved. This possibility is due to calculation of the cross section from the ratio of the number of ions N_i in the presence of saturation of the ion signal, i.e., under conditions in which Eq. (14) is satisfied, and the number of ions N_i^* for the same value of Q which would be observed at a much smaller value of τ when the condition $\int_0^\tau W(t)dt \ll 1$ is satisfied. The number of ions N_i^* is determined by extrapolation of data measured for $\int_0^\tau W(t)dt \ll 1$ in the region where saturation does not play an important role. In the presence of saturation the number of ions produced is related to the ionization probability not by Eq. (15) but by the relation

$$N_i = n \int \left[1 - \exp\left(-\int_0^\tau W(t)\, dt\right)\right] dv,$$

which in the case of the direct ionization process can be written in the form

$$N_i = nV_{k_0}\tau_{k_0}\alpha_{k_0}F_0^{k_0}\left[1-\frac{\alpha_{k_0}F_0^{k_0}\tau_{k_0}}{2!}\frac{V_{2k_0}}{V_{k_0}}+\frac{(\alpha_{k_0}F_0^{k_0}\tau_{k_0})^2}{3!}\frac{V_{3k_0}}{V_{k_0}}-\dots\right]. \quad (23)$$

In accordance with Eqs. (18)–(22) and (23), the ratio of the numbers of ions is

$$\frac{N_i}{N_i^*}=\left[1-\frac{\alpha_{k_0}F_0^{k_0}\tau_{k_0}}{2!}\frac{V_{2k_0}}{V_{k_0}}+\frac{(\alpha_{k_0}F_0^{k_0}\tau_{k_0})^2}{3!}\frac{V_{3k_0}}{V_{k_0}}-\dots\right]. \quad (24)$$

It is evident from Eq. (24) that with use of the relative method there is no need to measure the density of target atoms or to make absolute measurements of other quantities. It is just these facts which assure a significant increase in accuracy.

e) Laser radiation. Most experiments on multiphoton ionization of atoms have been carried out with radiation from pulsed Q-switched solid-state lasers, and also second harmonics of laser radiation. With a ruby laser the photon energy is $\hbar\omega \approx 1.78$ eV, and for a neodymium glass laser, ≈1.17 eV. In both cases the frequency of the radiation can be changed, in the case of the ruby laser within limits of ~10 cm^{-1} by changing the crystal temperature, and in the case of the neodymium glass laser within the width of the luminescence line, ~ 100 cm^{-1}, by using a dispersive resonator. Depending on the particular problem, radiation of definite polarization with various spectral widths and mode composition have been used.

In principle in those cases where a cross section must be measured, it is necessary to use the radiation of a laser operating with generation of one mode. Technical difficulties associated with achievement of stable single-mode operation have resulted in the use of a regime with generation of one transverse mode and many longitudinal modes. It is evident that with this type of radiation there will be no spatial fluctuations of intensity over the target, and the relations (20) are satisfied. Time fluctuations of intensity can be taken into account by introduction of a correlation function of k_0-th order. Studies of the generation of harmonics and multiphoton luminescence[70] for $k_0 \leq 3$ and of the multiphoton ionization of atoms[35] for k_0 from 5 to 11 have shown that in this case, when there are no nonlinear elements in the laser resonator, multimode radiation is satisfactorily described by a model of random phase distribution for which the correlation function is $\eta_{k_0} = k_0!$

f) Typical experimental arrangement (Fig. 3). The radiation of a laser 1 of a certain design is directed into a vacuum chamber 5. In front of the chamber in beam 2 is placed a series of light-splitter wedges 3 which separate auxiliary beams necessary for diagnostics of the laser radiation. The radiation spectrum is measured by means of a Fabry-Perot interferometer and a spectrometer 13. The energy in the laser radiation pulse is monitored by an auxiliary calorimeter 12. The time distribution of the radiation is measured by a coaxial photoelement 14. The spatial distribution of the radiation in the region of ion production is measured with an auxiliary objective 6′, identical to the objective 6 which focuses the light onto the target. The distribution of radiation in various planes of the focal region of the objective 6′ is imaged onto a photographic film through a microscope 15 with the necessary magnification. A radiation absorber 4 of a certain type is placed in the beam in front of the vacuum chamber. The radiation energy which has passed through the focal region 7 is measured by a calorimeter 11 placed beyond the chamber. The chamber is filled with the gas being studied or an atomic beam is sent into it. In a direction perpendicular to the beam of light are placed the accelerating field electrodes 8, the electron optics which focuses ions onto the detector, a flight-path interval, an electron multiplier 9, and a Faraday cup 10, which is used in the case where it is necessary to measure the absolute number of ions.

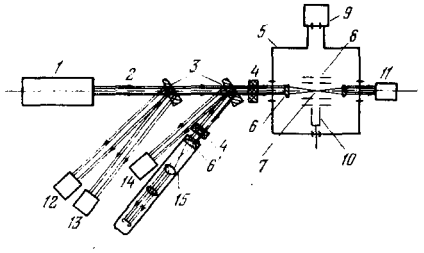

FIG. 3. Diagram of typical experiment to study multiphoton ionization of atoms (see the text).

Observation of multiphoton ionization of the gas studied is established by detection of the ion signal due to admission of gas or turning on the atomic beam and having a given delay time relative to the laser pulse.

An experiment to measure the degree of nonlinearity of the process consists in changing the attenuation of the radiation at the entrance to the chamber, and measuring the energy passing through the region of focusing and the corresponding amplitude of the ion signal in relative units. A typical form of a set of experimental data obtained in a series of consecutive laser radiation pulses is shown in Fig. 4 on a doubly logarithmic scale. The experimental data have been approximated by a straight line obtained by the method of least squares. The upper limit of the region of approximation was systematically shifted toward the region of higher intensities until the quantity $k = \partial \log A_i / \partial \log Q$ remained constant. Thus, we separated the region of variation of intensity in which the ion signal undergoes saturation due to achievement of a significant degree of ionization in the region of ion production (see Eq. (23)).

An experiment to measure the cross section for multiphoton ionization of an atom consists of the simultaneous measurement of all quantities entering into Eq. (21). At the same time the necessary parameters of the laser radiation are monitored (the frequency at the peak of the radiation spectrum, the width of the spectrum, and the polarization and mode composition of the radiation).

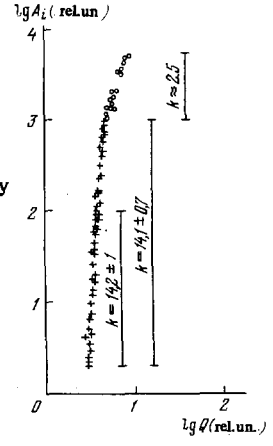

FIG. 4. Typical experimental result on multiphoton ionization of atoms—dependence of ion signal pulse height A_i on energy in laser pulse passing through the target Q, in a log-log plot. The region where saturation of the ion signal arises as the result of achievement of a significant degree of ionization is evident (k ≈ 2.5, data of ref. 11).

TABLE V. Values of various parameters realized in measurement of the cross section α_5 for direct ionization of the sodium atom (f and d are the focal length and relative aperture of the planoconvex lens used to focus the laser radiation; D is the cross-section diameter of the light beam at the entrance aperture of the focusing lens; the remaining designations are given in the text)

k_0	Q, J	τ_1, nsec	τ_b, nsec	f, mm	d, mm	D, mm	S, cm^2	V_1, cm^3	V_b, cm^3	N_i	n, cm^{-3}
5	10^{-2}	30	15	100	10	3	10^{-5}	$5\cdot10^{-6}$	10^{-6}	10^3	10^9

In Table V, as a quantitative illustration of typical experimental conditions, we have shown some values obtained in measurement of the cross section for direct five-photon ionization of the sodium atom by radiation from a neodymium glass laser.[36]

In ending our review of experimental arrangements, we must make three remarks. The first is that, beginning with the first experiments and continuing up to the present time, the main difficulty consists of maintaining constant and measuring the space-time distribution of radiation over the target; the requirements increase rapidly as the degree of nonlinearity of the process studied increases. Second, it is obvious that an optimum experiment is carried out in a single-mode laser. Third, there is unquestioned promise for dye lasers, which already permit studies to be made of few-photon processes practically over the entire range of optical frequencies.

4. DIRECT MULTIPHOTON IONIZATION

a) Conditions of achievement. The condition for achievement of direct ionization, expressed in terms of the difference between the energy $1 \leq k_0 \leq k_0 - 1$ of the photons and the energies E_i associated with the electron states in the unperturbed spectrum of the atom, is of the form

$$|k\hbar\omega - E_{0i}| > \delta E_{0i}(\mathscr{E}),\ \gamma_{0i}(\mathscr{E}). \qquad (25)$$

Here E_{0i} is the tabulated energy of the transition between states 0, i in the spectrum of the atom in the absence of a field, $\delta E_{0i}(\mathscr{E})$ is the change of the transition energy under the action of the field, and $\gamma_{0i}(\mathscr{E})$ is the reduced width of the states 0, i in a light field. It should be noted again (see Chap. 1) that in the general case in a varying field it is impossible to neglect the change of energy of the ground state 0 in comparison with the excited state i. On the other hand, in most cases, where $k_{iE} \ll k_{0E}$, we can neglect the broadening of the ground state 0 in comparison with the broadening of the state i.

The condition (25) is essentially the condition of a large energy difference in the absence of a field and weak perturbation of the atomic levels by the field. Achievement of a large energy difference is facilitated at a high frequency of the light and by a small excess of the energy $k_0\hbar\omega$ over the ionization potential I. Under these conditions the energy $k\hbar\omega$ will be maximally removed from the upper, closely spaced levels. To achieve the condition of weak perturbation of the atomic levels, it is most important that the field be relatively weak. Finally, even in an arbitrarily weak field resonances may appear, but for this a special choice of radiation frequency is necessary. In a strong field or for an energy $k\hbar\omega$ corresponding to the energy of the closely spaced upper levels, the probability of achieving the relation (25) is decreased.

Finally, we note that in practice an additional quantity Γ, taking into account the nonidealness of the experimental arrangement, must be added to the right-hand side of the inequality (25). Depending on the specific conditions, Γ is determined by the Doppler broadening of the atomic levels and by the inhomogeneity broadening associated with the nonuniform distribution of the light field over the target and with the nonmonochromaticity of the laser radiation.

b) Observation of the direct process. The direct process of multiphoton ionization has been observed over a wide range of variation of the degree of nonlinearity $2 \leq k_0 \leq 11$, of field strength 10^5–5×10^7 V/cm, and adiabaticity parameter $1000 \geq \gamma \geq 5$ in ionization of various atoms (hydrogen, the alkali atoms, mercury, the noble gases) from the ground and excited states by light with various wavelengths and degrees of polarization (Table VI). A strict criterion of occurrence of direct ionization is the constancy of the power dependence W(F) for a small variation of the radiation frequency, where the quantity $k\hbar\omega$ remains in the intervals between resonances. However, under conditions where the energy differences in the absence of a field are large and the calculations permit the assumption that the perturbation of quasiresonance states is small, the criterion may be the power dependence W(F) measured for a fixed radiation frequency. These conditions were satisfied in most cases and the results of the investigations are listed in Table VI. In a number of experiments the frequency was varied as a control.[36,53] This type of control was achieved also in experiments carried out in a relatively weak field[44], where the theoretical results are inapplicable. It is necessary to note specially the observation of the direct process for $\gamma \approx 5$ (the Xe atom, $\hbar\omega = 1.17$ eV, $\mathscr{E} = 5 \times 10^7$ V/cm). Analysis of the conditions of this experiment was carried out by Alimov[59] on the basis of general considerations regarding the ionization probability for $\gamma \sim 1$ obtained in refs. 1 and 28. It turned out that, although at $\gamma \sim 5$ the probabilities for ionization as the result of absorption of k_0, $k_0 + 1$, $k_0 + 2$, ... photons already have comparable values, nevertheless for the integral ion yield the deviations from a power law (1) lie within the experimental accuracy in measurement of the value of k. Therefore, although the observation $k \approx 11 = k_0$ in certain intervals of variation of the radiation frequency shows that the ionization process has a direct nature, we must assume that the calculations, which were carried out by the method

TABLE VI. Experimental and theoretical data on direct multiphoton ionization with linear polarization of the radiation (the dimensionality of the multiphoton cross section is $[\alpha_{k_0}] = \text{cm}^2 k_0 \text{sec} k_0^{-1}$)

k_0	2	3		4	5	6			7
Atom	K	Na	Cs	K	Na	H	Hg	Kr	Xe
$\hbar\omega$, eV	2.36	2.36	1.78	1.18	1.18	2.36	1.78	2.36	1.78
$\mathscr{E}$, V/cm	10^5	$5\cdot10^5$	$5\cdot10^5$	10^6	$5\cdot10^6$	10^7	10^7	10^7	10^7
γ *	10^3	$5\cdot10^2$	$5\cdot10^2$	10^2	25	40	30	40	30
k_{exp}	1.8	3.0	3.0	4.0	4.9	5.7	6.3	5.7	7.4
Δk_{exp}	±0.2	±0.2	±0.2	±0.1	±0.1	±0.5	±0.7	±0.6	±0.8
$\lg(\alpha_{k_0})_{coh}$	—49.1	—80.4	—77.8	—108.4	—140.1	—179.7	—174.4	—173.0	—207.4
$\Delta \lg(\alpha_{k_0})_{coh}$	±0.8	±1.0	±1.5	±1.4	±1.7	±1.8	±2.3	±2.0	±2.8
Reference	36	36	38	36	36	40	41	42	41
Bebb's method [14]	—48.5	—79.2	—76.0						
Gold's method [16]						—180.1			—210.1
Morton's method [17]	—48.6	—77.5	—76.0	—108.5	—140.3	—181.4	—174.9	—181.2	—211.6
Green's function method [21]	—48.8	—77.7	—75.0	—106.4	—137.4				
The same, [20]						—179.7			

*Approximate values are given which correspond to the maximum field strength for which the power law (1) is observed.

of standard perturbation theory, should not directly describe the probability value.

In concluding our discussion of the conditions under which the direct process is observed, it is necessary to make the trivial but important remark that there is no single limit of the field strength making possible realization of the direct process. For each specific atom and each radiation frequency there are always specific differences between the energies $k\hbar\omega$ and the energies of the stationary bound states, a specific nature of the perturbation of these states, and a specific field strength at which resonances induced by the field arise. It is evident that for resonances induced by the field arise. It is evident that for resonance frequencies where $k\hbar\omega = E_{0i}$ (where E_{0i} is the energy of the transition the quasi-stationary state i), the ionization process will have a resonance nature in an arbitrarily weak field. A particular case of resonance situation may be an energy $k\hbar\omega$ very close to the limit of the spectrum of bound states. In this case a role can be played by difference effects whose role has not yet been completely clarified–formation of a continuum as the result of nonresonance mixing of upper, closely spaced bound states, increase of the ionization limit as the result of oscillational energy of the electron in the field of the wave, and threshold phenomena.

c) Direct-ionization cross sections. In Table VI we have given the cross sections for direct multiphoton ionization processes measured with linear polarization of the light. In all cases the data were obtained by an absolute method of measurement of the multiphoton cross sections (see Chap. 3). Since the laser radiation had a multimode nature, the cross section for single-mode radiation $(\alpha_{k_0})_{coh}$ was determined from the value measured in multimode radiation α_{k_0} by the relation:

$$(\alpha_{k_0})_{coh} = \frac{1}{\eta_{k_0}} \alpha_{k_0} = \frac{1}{k_0!} \alpha_{k_0}.$$

The experimental results given in Table VI are the most reliable ones. In addition to these data, other measurements have also been published of a number of cross sections (for example ref. 39), but these have not been listed because of their lower experimental reliability.

Several cross sections have been measured with significantly higher accuracy achieved as a result of using the relative-measurement method and single-mode radiation (Table VII).

The dependence of the cross section on polarization of the radiation has been investigated experimentally by observation of the ratio of amplitudes of the ion signals for linear and circular polarization. The polarization of the initial (98–99%) linearly polarized laser radiation was varied by changing the orientation of a $\lambda/4$ plate placed in the light beam between the laser and the target. On varying the polarization of the radiation by this method the spatial distribution of the radiation in the beam remained constant, and therefore the ratio of amplitudes of the ion signals corresponded to the ratio of the cross sections. The results of these measurements are shown in Table VII, together with the theoretical results. It should be noted that measurements of the ratios of the cross sections for light of different polarizations permit obtaining data significantly more accurate than measurements for a fixed polarization carried out by an absolute method. The calculations of the cross sections for the direct ionization process in a circularly polarized field were carried out with a Green's function constructed by the quantum defect method.[21]

TABLE VII. Cross sections measured experimentally by a relative method

k_0	3	4	5
Atom	K		Na
$\hbar\omega$, eV	1,78	1,17	1,17
Radiation	Single-mode	Multimode	Single-mode
lg (α_{k_0}) coh	−78.5	−109.0	−136.5
Δ lg (α_{k_0}) coh	±0.1	±0.2	±0,5
Reference	45	34b	35b
Bebb's method [16]	−79.3		
Morton's method [17]	−78.1	−108.5	−140.3
Green's function method [21]	−78.7	−106.4	−137.4

The dependence of the ion yield on the polarization of the light has been measured also for the intermediate case for $\gamma \sim 5$ (the Xe atom, $k_0 = 11$).[59] The result is shown in Fig. 5 in the form of the ion yield as a function of the degree of ellipticity of multimode radiation for a fixed average field strength. Since at the present time there are no data on the effective correlation factor for a nonpower function $W(F)$ on the dependence of this factor on the degree of polarization, there is no possibility of deducing from the experimental data the dependence of the ionization probability on the ellipticity of the light.

The cross sections calculated by standard perturbation theory are also given in Tables VI–VIII. Analysis of the conditions of appearance of resonances induced by the field, as carried out in Chap. 5, shows that for a field strength $>10^7$ V/cm the spectrum of bound states as a rule is strongly perturbed by the field, and although it is always possible to find a frequency at which the ionization has a direct nature, it is nevertheless unknown what effect the change of basis has on the value of the cross section. Therefore adequately justified conclusions can drawn only from data obtained for field strengths $\lesssim 10^6$ V/cm, i.e., for relatively few-photon processes. If we turn to these data, then as can be seen from Tables VI-VIII the calculations by standard perturbation theory satisfactorily describe the experimental results. It is also evident from Table VII that the cross-section measurements carried out by the relative method permit attainment of an accuracy sufficient for determination of the optimal method of calculation.

In conclusion it should be noted that the problem of creating good methods for describing the direct multiphoton ionization of atoms arises not only from the

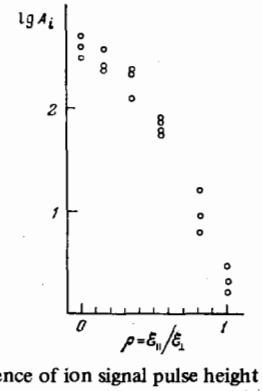

FIG. 5. Dependence of ion signal pulse height A_i (~ionization probability) on radiation ellipticity $\rho = \mathcal{E}_\parallel / \mathcal{E}_\perp$. The eleven-photon ionization of Xe at a field strength of $\sim 5 \times 10^7$ V/cm ($\gamma \approx 5$, data of ref. 59).

TABLE VIII. Measurements and calculations of the ratio of the cross sections for direct multiphoton ionization by light of linear polarization ($\alpha^L_{k_0}$) and circular polarization ($\alpha^C_{k_0}$). The ratios are given for a fixed average value of field strength.

k_0	2		3			4	5
Atom	K	Cs	Na	K	Cs	K	Na
$\hbar\omega$, eV	2.36	3.56	2.36	1.78	1.78	1.18	1.18
$\alpha^L_{k_0}/\alpha^C_{k_0}$	1.0	0.8	2.4	0.4	0.5	0.6	2.0
Reference	$\pm^{0.85}_{46}$	$\pm^{0.1}_{37}$	$\pm^{0.5}_{46}$	$\pm^{0.1}_{45}$	$\pm^{0.1}_{37}$	$\pm^{0.2}_{46}$	$\pm^{0.4}_{46}$
Theory [21]	0.85	0.9	3.0	0.4	0.7	0.25	12

natural desire to learn the unknown laws of nature, but also is dictated by practical requirements, since the task of experimental tabulation of cross sections is obviously insoluble.

d) Main results of investigations. In summarizing the results of experimental and theoretical research on the direct multiphoton ionization of atoms, it is necessary first of all to mention that for $\gamma \gg 1$ the direct process is observed for a wide variation of the various parameters describing the field and the atom. Comparison of the entire set of data on cross sections with the theoretical results permits us to draw the optimistic conclusion that standard perturbation theory satisfactorily describes the process, in any case up to a field strength of 10^7 V/cm. However, it should be noted that the optimism of this conclusion may be due to the frequencies at which the measurements have been carried out, far from the characteristic frequencies (resonances, minima) and located in the region where the cross section is not a strong function of the specific structure of the atomic spectrum. A more accurate answer can be given by investigation of the frequency dependence of the ionization probability in a circular field in the region where standard perturbation theory predicts existence of interresonance minima. Observation of these minima presents independent interest as the observation of bands of transparency of an atomic medium for nonlinear absorption of light.

The question of the upper limit of field strength to which calculations carried out by means of standard perturbation theory are applicable, as mentioned above, has no universl answer. The limit depends substantially on the initial differences and the amplitude of perturbation of the closest levels. In the case in which the differences are large and change insignificantly on turning on of the field, the limit should be determined only by the satisfaction of the condition $\gamma \gg 1$. The entire set of experimental data confirms this conclusion.

Finally, we turn to the direct process occurring for an adiabaticity parameter $\gamma \sim 1$. However, a trivial remark must first be made: for $\gamma \sim 1$ we can discuss only measurement of the probability for a fixed field strength, since the power law (1) is not satisfied and there is no constant which does not depend on the field strength. A theoretical analysis of the ionization process for $\gamma \sim 1$, carried out in ref. 28 in the quasiclassical approximation, showed that the degree of polarization of the radiation substantially affects the probability, since the distribution in the number of absorbed photons is different for light of different polarizations. As mentioned above in Chap. 2, an exact theoretical calculation has been carried out for $\gamma \sim 1$ for circularly polarized light.[28a] Unfortunately, all of the experimental data for the direct process for $\gamma \sim 1$ have been obtained in multimode radiation. Comparison of these data with the theory is further hindered by the absence of information on the correlation factors for a nonpower dependence of the ionization probability on radiation intensity. Therefore it is important both to calculate the correlation factor and to perform experiments for $\gamma \sim 1$ with use of single-mode radiation. In this way it is possible to establish conditions in which the channel of electron penetration through the potential barrier begins to appear.

5. RESONANCE MULTIPHOTON IONIZATION

As we have mentioned above, the multiphoton ionization process is called resonance in the case in which an intermediate resonance arises between the energy of $k < k_0$ photons of the radiation and the energy of the electron transition from the ground state to some excited bound state of the system atom + strong light field. Therefore it is evident that the condition of realization of the resonance ionization process is the opposite of condition (25) for realization of the direct ionization process; it has the form

$$|k\hbar\omega - E_{0s}(\mathscr{E})| < \gamma_{0,s}(\mathscr{E}), \tag{26}$$

where $E_{0s}(\mathscr{E})$ is the energy of the transition from the ground state 0 to the state s in the system atom + light field, and the remaining designations are the same as above. In regard to the quantities $E_0(\mathscr{E})$, $E_s(\mathscr{E})$, $\gamma_0(\mathscr{E})$, and $\gamma_s(\mathscr{E})$ it is necessary to make the same remarks which have been made in analysis of the condition (25) for realization of the direct process. The natural desire arises to describe the condition for the resonance ionization process in terms of the quantities which characterize the unperturbed spectrum of the atom. Unfortunately, this cannot be done in general form; specific data on the perturbation of the resonance states are necessary.

Equation (26) is a necessary but not sufficient condition for realization of a 0s resonance transition in a strong light field. In addition to satisfying (26), it is necessary that the 0s transition be allowed by the selection rules for multiphoton transitions. Depending on the degree of polarization of the radiation, the projection of the orbital angular momentum of an electron which has absorbed several photons differs substantially–the projection has a value 0, ±1 for linear polarization and a value k_0 for circular polarization (all photons have identical helicity!). A large value of the quantum number l requires a correspondingly large value of the principal quantum number n which characterizes the state s.

Realization of a 0s resonance transition can lead both to multiphoton excitation of the atom and to ionization of the atom if an intermediate resonance exists. These processes are competitive. For dominance of the multiphoton excitation process, it is necessary that the probability of the s0 transition be greater than the probability of the transition sE from the state s to the continuum. This relation between the probabilities is satisfied if the degree of nonlinearity of the 0s transition is less than the degree of nonlinearity of the transition sE ($k_{0s} < k_{sE}$) and no saturation occurs in the first transition.

Let us turn to the resonance ionization process, which is always dominant for $k_{0s} > k_{sE}$. As already pointed out in chapter 2, in the language of perturbation theory the appearance of an intermediate resonance signifies a sharp decrease in the corresponding energy

denominator in the compound matrix element (10) to the value determined by the width of the resonance state $\gamma_S(\mathscr{E})$. This permits, with sufficient accuracy for description of the resonance process, taking into account only this resonance term in Eq. (10). With use of one resonance term it is possible to describe correctly the frequency dependence of the probability of resonance ionization, since the discarded nonresonance terms are weak functions of frequency. The ionization probability in this case is related to the parameters describing the resonance state[1,24a] as follows:

$$W \sim \frac{\mathscr{E}^{2k_0}}{[E_{0s}(\mathscr{E}) - k\hbar\omega]^2 + [\gamma_{0s}(\mathscr{E})]^2}. \qquad (27)$$

It should be noted that, as mentioned above, $\gamma_{0S}(\mathscr{E}) \approx \gamma_S(\mathscr{E}) = W_{SE}$. With use of Eq. (27) for quantitative analysis of the experimental data, it is necessary to have in mind that there is no general solution of the problem of a transition through an intermediate resonance state to the continuum, which includes consideration of the competition of the direct and cascade transitions and which does not neglect the perturbation of the resonance state by the radiation field. Individual special cases have been considered in refs. 1 and 24.

a) **Experimental methods.** In investigation of the resonance ionization process it is necessary to take into account three instrumental factors—the width of the laser radiation line and the Doppler and inhomogeneity broadenings of the resonance state. A first trivial remark is that, the smaller these widths, the sharper the resonance appears. Second, it must be noted that there is a dependence of the reduced width of the radiation line exciting the multiphoton resonance Γ_k on the degree of coherence of the radiation. Thus, in the case of single-mode radiation $\Gamma_k = k\Gamma$, and in the case of multimode radiation with a random phase distribution $\Gamma_k = \sqrt{k\Gamma}$ (where k is the degree of nonlinearity of the resonance and Γ is the width of the radiation line). Third and finally, we must note the important role of the inhomogeneity broadening. If the line width of the laser radiation can be decreased by many orders of magnitude to 10^{-4} cm^{-1} (in the case of single-mode lasing), then the space-time distribution of the light over the target is always nonuniform and it is impossible to change this degree of nonuniformity substantially. The effect of the nonuniformity reduces qualitatively to the fact that the principal fraction of the ionized atoms are in some effective field which, in spite of the strongly nonlinear nature of the ionization process, is always substantially less than the maximum value (since only a small fraction of the neutral atoms are in the region where the field is maximal). Quantitative allowance for the nonuniformity of the distribution[48,51] shows that its role decreases sharply as the degree of nonlinearity of the resonance increases.

1) Observation of frequency dependence of ion yield for fixed field strength. An intermediate resonance appears in the resonance rise of the ion yield as the frequency of the laser radiation changes (Fig. 6); the energy of the transition to the resonance state is determined from the maximum in the yield curve with allowance for the inhomogeneity broadening. If the reduced width of the radiation line is $\Gamma_k < \gamma_S(\mathscr{E})$, then the half-width of the resonance state is determined by the half-width of the maximum in the ion yield with allowance for the inhomogeneity broadening. To determine the dependence of the transition energy to the resonance state on the field strength, it is necessary to make a number of measurements at different field strengths (see Fig. 6). The main practical difficulty in using this method is the necessity of maintaining the field strength constant as the radiation frequency varies.

2) Observation of dependence of ion yield on field strength for fixed radiation frequency. The appearance of an intermediate resonance is identified from the resonance deviation of the function $A_i(Q)$ from the power law describing the direct ionization process (Fig. 7). A resonance will appear if the condition $\Delta E_{0S}(\mathscr{E}) > \gamma_{0S}(\mathscr{E})$, Γ_k is satisfied in the interval of variation of the field strength used. The energy of the transition to the resonance is determined by the frequency at which resonance is observed with allowance for the inhomogeneity broadening. Determination of the dependence of the transition energy on the field strength requires performance of a series of measurements of the function $A_i(Q)$ at different radiation frequencies (Fig. 7). The main deficiency of the method is the necessity of varying the field strength over wide limits, which is possible only in the case of relatively few-photon processes.

3) Observation of dependence of $k = \partial \log A_i / \partial \log Q$ on radiation frequency. In this method the resonance is identified from the deviation of the experimentally observed value of k from the corresponding quantity k_0. The quantity k is an integral characteristic of the ionization process and its determination requires measurement of the ionization probability as a function of field strength over some interval of variation of the field strength. The occurrence of the resonance is also determined substantially by the field strength. Therefore this method is applicable only under conditions where the range of variation of field strength necessary for measurement of k is small in comparison with the value which excites (or disrupts) resonance. If this condition is satisfied, the nonpower function $A_i(Q)$ can be approximated with sufficient accuracy by a power law in a definite interval of variation of Q. The observation of $k \neq k^0$ can serve as an indication of the existence of resonance. Variation of k is produced by the change in the transition energy under the influence of the field for the condition $\Delta E_{0S}(\mathscr{E}) \geq \Gamma_k, \gamma_{0S}(\mathscr{E})$. If the resonance is detuned under the influence of the field, we should observe $k < k_0$, and if it is excited, then we should observe $k > k_0$, since simultaneously with the increase of the intensity of the radiation there is a decrease (or an increase) in the number of photons on absorption of which a resonance transition occurs. For a fixed frequency of light, the observed value of k depends on the initial detuning of the resonance. In Fig. 8 we have shown the results of calculation of the value of k for the case of three-photon resonance in four-photon ionization. The two limiting cases, $\Delta E_{0S}(\mathscr{E}) \lesssim \gamma_{0S}(\mathscr{E})$, for which the calculation was made have been observed experimentally.[53,54] If $\Delta E_{0S}(\mathscr{E}) > \gamma_{0S}(\mathscr{E})$, Γ_k, then the point of intersection of the curve $k(\omega)$ with the axis of abscissas (see Fig. 8) corresponds to the frequency at which resonance is observed for the field strength realized in the experiment. A practical deficiency of the method is the necessity of accurate measurement of the function $A_i(Q)$ over a narrow interval of variation of the radiation intensity. The value of $\gamma_{0S}(\mathscr{E})$ can be determined only from comparison of the experimental data and theoretical calculations.

In conclusion it should be noted that the value of k observed experimentally in the presence of an inter-

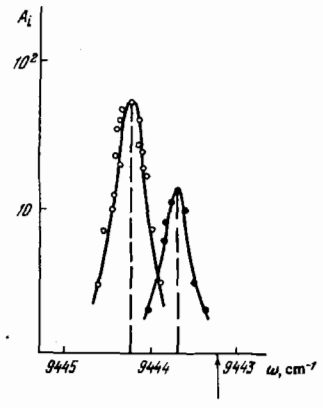

FIG. 6

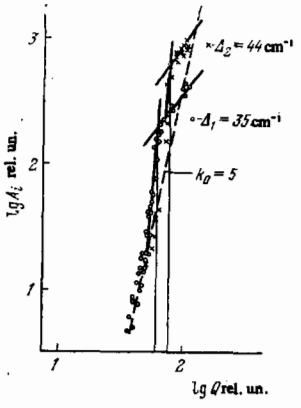

FIG. 7

FIG. 6. Observation of resonance multiphoton ionization by means of the frequency dependence in the ion yield $A_i(\omega)$. The three-photon resonance 6s–6f in four-photon ionization of the Cs atom. Resonance has been observed for two values of field strength related as 1.0: 1.5. The arrow shows the frequency corresponding to the three-photon resonance 6s–6f in the unperturbed spectrum of the atom (data of ref. 48).

FIG. 7. Observation of resonance multiphoton ionization by means of the dependence of the quantity $k = \partial \log A_i / \partial \log Q$ on radiation intensity (frequency fixed). The four-photon resonance $2^3s - 14^3s$ in the five-photon ionization of metastable helium. $\Delta_{1,2}$ are the detuning of the resonance in the absence of a field (data of ref. 52a).

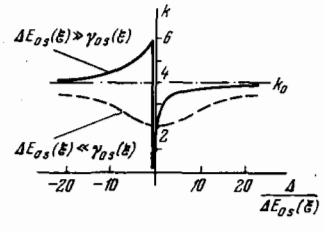

FIG. 8. The quantity $k = \partial \log W / \partial \log F$ (W is the ionization probability, F is the radiation intensity) as a function of the frequency difference $\Delta$of the resonance in the absence of a field, the change in transition energy $\Delta E_{0s}(\mathcal{E})$, and the width of the states $\gamma_{0s}(\mathcal{E})$ under the action of the field. Calculation for the intermediate three-photon resonance in four-photon ionization [51].

mediate resonance is not directly connected either with the degree of nonlinearity of the ionization process or with the degree of nonlinearity of the resonance excitation process.

b) Results of investigations of the resonance ionization process. Resonance multiphoton ionization has been observed experimentally in many cases–for various atoms, for linear and circular polarization of the light, and over a wide range of field strength and value of the parameter γ. All of the experimental results can be separated into two groups. The first group comprises the results which it has been possible to interpret as appearance of an intermediate resonance with a definite electronic bound state. As a rule, this possibility has been due to the relatively low degree of nonlinearity of the ionization process observed, corresponding to relatively low field strength and small perturbation of the resonance state. In most cases the change in the transition energy observed in the experiment could be quantitatively described by calculations carried out by perturbation theory. The second group comprises resonances which it has not been possible to identify. These represent the case of a high degree of nonlinearity and high field strength, where a strong perturbation of the resonance state is observed and, correspondingly, perturbation theory is not applicable to description of the spectrum of the system atom + light field.

1) Identified resonances. An example of resonance ionization observed in such a weak field that the perturbation of the resonance state can be neglected is the three-photon ionization of the cesium atom in the presence of intermediate two-photon resonances from the ground state to highly excited s and d states having principal quantum numbers[47] $n \approx 9–13$. The radiation from a dye laser was used for the ionization, and the frequency was varied over the range 6550–6950 Å with a line width 0.06–0.08 Å. The ionization was observed for a field strength $\sim 10^4$ V/cm. A typical result is shown in Fig. 9. Note the red shift of the $D_{5/2,3/2}$ doublets. The main value of such experiments lies in the possibility in principle of measuring the matrix elements of bound-bound transitions, including transitions forbidden by the selection rules for single-photon absorption (an example is the transitions observed in ref. 47). For this purpose, however, it is necessary not only to carry out absolute measurements of the ionization probability in the resonance, but also to take into account carefully all factors determining the width of the resonance state. In spite of various difficulties encountered in determination of the width (some of these were pointed out above), the development of research in this area appears very promising.

An example of observation of the case in which the atomic spectrum is substantially perturbed by the field is the four-photon ionization of the cesium atom in the presence of the intermediate three-photon resonance[48] 6s-6f. In this experiment the ionization was accomplished with a single-mode neodymium glass laser with variable frequency. The frequency dependence of the ion yield was measured for a fixed field strength. Absolute measurements of the field strength permitted determination of the constant in the square law for variation of the transition energy (Table IX). The value of the constant is given with satisfactory accuracy by calculations taking into account among other things the nonresonance mixing[7] of the components of the 6f doublet in the field. This method has been successfully used also in a number of other experiments (the two-photon resonance 2^1s-6^1s for the three-photon ionization of metastable helium[49]; the 4s-6s two-photon resonance in the three-photon ionization of potassium[50]). As a whole the results show high reliability of these measurements. However, measurements at a series of values of field strength can be accomplished only for a small degree of nonlinearity of the ionization process.

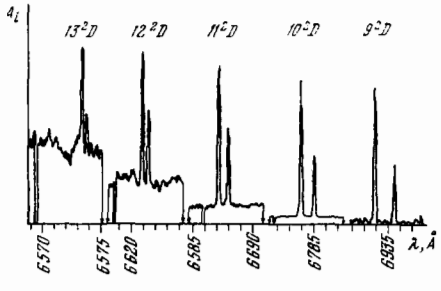

FIG. 9. Three-photon resonance ionization of the cesium atom in the presence of intermediate two-photon resonances with the states indicated in the figure (ground state 6s, data of ref. 47).

TABLE IX. Measurement of perturbation of atomic levels in a light field (α is the constant for the quadratic variation of the transition energy with the field; k is the degree of nonlinearity of the resonance transition; the approximate field strength at which the measurements were made is given)

Atom	Transition	k	λ, μ	V/cm	α, $\frac{cm^{-1}}{GW/cm^2}$	Method	Reference
Hg	$7^3s_1 - 6^3p_2$	—	1.06	10^5	10	Observation of relaxation	4c
K	$4p_{3/2} - 6s_{1/2}$	—	0.69	10^5	10	Absorption of light of auxiliary source	4a
He	$2^1s - 6^1s$	2	0.69	10^5	15	Resonance multiphoton ionization	49
He	$2^3s - 13^3s$	4	1.06	10^6	60	The same	52b
He	$2^3s - 14^3s$	4	1.06	10^6	150	» »	
He	$2^3s - 14^3d$	4	1.06	10^6	150	» »	
Cs	$6s - 6f$	3	1.06	10^6	1.5	» »	48

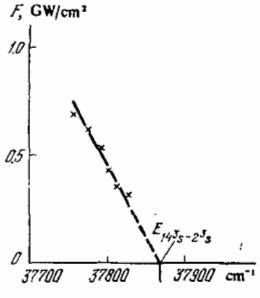

FIG. 10. Energy of $2^3s - 14^3s$ transition as a function of radiation intensity. The transition was observed as the intermediate four-photon resonance in five-photon ionization of metastable helium. The result of a number of experiments, data of two of which are given in Fig. 7 (data of ref. 52b).

The method of changing field strength for a fixed frequency of radiation has also been used successfully. Studies[52] of the five-photon ionization from the metastable state 2^3s of the helium atom by linearly polarized radiation from a neodymium laser have permitted data to be obtained on a large number of resonance processes. With a small change of the laser frequency, the energy of the four photons can be made equal to the energy of the transition from the 2^3s state to the s and d states having principal quantum numbers 13–15. The experiment observed the dependence of the ion yield on the radiation intensity for a number of fixed frequencies. (A typical result is shown in Fig. 7.) In each case the frequency and intensity of the radiation corresponding to resonance deviation in the ion yield was measured. The entire set of observed resonances was recorded in coordinates ω and F. In a number of cases the experimental points lay on a straight line whose extrapolation to the value F = 0 gave an energy corresponding to the energy of a definite transition in the unperturbed spectrum of the helium atom. An example is the data for the 2^3s-14^3s transition, which are shown in Fig. 10. For a large number of closely spaced highly excited levels with which resonances could arise and with a large change of the transition energy in the field, this method of identification of a resonance transition has turned out to be rather reliable. A quadratic variation of transition energy with field strength has been observed for a number of resonance ionization processes. The values of the constants measured in these experiments are given in Table IX. (In interpretation of the data in Table IX and in Fig. 10, it is necessary to bear in mind that what is measured experimentally is the result of perturbation of both states, initial and resonance, and that the magnitudes of these perturbations depend in a different way of the radiation frequency, so that it cannot be predicted a priori which of them will dominate.) This method has been used also in a number of other experiments which, however, did not yield very reliable results (for example ref. 55), since sufficiently detailed measurements with variation of both parameters ω and F were not carried out.

In a number of experiments the resonance ionization process has been identified by observation of deviation of the quantity $k = (\partial \log A_i)/(\partial \log Q)$ from the corresponding quantity k_0 (refs. 43, 44, 51, 53, 54, and 57). As mentioned above, interpretation of the results obtained by this method is difficult. Therefore the use of this method does not as a rule give any data other than the conclusion that an intermediate resonance exists.

In Table IX we have listed the data of a number of experiments in which resonance ionization was observed and in which it was possible to measure the change in the transition energy in the atomic spectrum under the influence of a strong light field. All of the data were obtained for linear polarization of the radiation. For comparison we have listed also typical data obtained by classical methods. It is important to note that the variation of the transition energy for two-, three-, and four-photon excitation in a field of strength up to 10^6 V/cm is quadratic in the field. The large spread in the magnitude of the data obtained should not be surprising. We recall that in a variable field the amplitude of the perturbation is determined not only by the principal quantum number of the state, but also by the radiation frequency (see Eq. (6)).

It should, however, be noted that in a number of experiments, for example, in the same experiments[52] on ionization of metastable helium, resonances not described by a quadratic law have been observed. The cause of the observed phenomena is not yet known. Therefore a continuation of studies of the resonance process in not very strong fields is an important goal.

Measurement of the perturbation of an atomic spectrum in a light field of intensity up to 10^6 V/cm and especially data for highly excited states present exceptional interest for understanding of the phenomena which determine the spectrum of the system atom + light field.

2) Resonances in an elliptically polarized field. The main interest is due to the possibility of observing resonances with quasistationary states arising[10b] on perturbation of the atomic spectrum by an elliptical field. Unfortunately, no such data have been obtained as yet. Only the dependence on the degree of ellipticity due to the selection rules has been observed. Thus, Bakos[49] in accordance with the selection rules observed the two-photon resonance 2^1s-6^1s in helium only in a linearly polarized field and did not observe it in a circularly polarized field. In agreement with the selection rules, the two-photon resonances in potassium[50] in a linear field 4s-6s and in a circular field 4s-4d have been observed. These two experiments were carried out at a field strength $\sim 10^5$ V/cm. The role of polarization of the light turns out to be important also in a very strong field[59]–the resonance observed in the eleven-photon ionization of xenon in a field $\sim 5 \times 10^7$ V/cm in the case of linear polarization is not observed with circular polarization (see below). Experimental verification of the satisfaction of selection rules for single-photon processes is of definite interest, especially in the case of high field strength where the ratio between the probabilities of forbidden and allowed transitions can change. However, the main interest is nevertheless in the experimental observation of the change in the spectrum in an elliptical field.

3) Resonance ionization at high field strength. Unidentified resonances. Studies of the frequency dependence of highly nonlinear multiphoton ionization of noble gas atoms, observed at field strengths greater than 10^7 V/cm, have shown that the nature of the process changes considerably for an insignificant change in frequency. Such data have been obtained for atoms of xenon[44,57,59], krypton[57], and neon[56]. Both a change in the value of k and a change in the yield of ions have been observed for a change in frequency by ~1%. The eleven-photon ionization of the xenon atom by radiation from a neodymium glass laser, $\hbar\omega \approx 9400$ cm^{-1}, has been studied in greatest detail.[44,57,59] The laser frequency was varied over ≈100 cm^{-1} for a line half-width of about ≈1 cm^{-1}. Ionization was observed in linear and circular fields for a field strength of about 5×10^7 V/cm. In the case of circular polarization of the radiation it turned out that the nature of the ionization process did not depend on the frequency of the light, and the ionization probability is related to the intensity of the radiation by an expression of the form $W \sim F^{11\pm1}$, i.e., the ionization process has a direct nature. With linear polarization in the interval 9400–9430 cm^{-1} the ionization process has a direct nature, and in the interval 9430–9460 cm^{-1} it has a resonance nature (Fig. 11). Since the energy of the transition to the first excited state in the xenon atom is ~8$\hbar\omega$, a resonance can arise in absorption of nine or ten photons. In both cases the transition energy to the resonance state, corresponding to the observed maximum in the ion yield for $\hbar\omega_{res} = 9445$ cm^{-1}, differs from the energy of the allowed transitions in the absence of a field by an amount ~100 cm^{-1}. The same scale of values applies to the distances between levels in the spectral region where a resonance can appear. Under such conditions the perturbation of the spectrum cannot be considered small, so that there is no basis for use of standard perturbation theory either for calculation of the perturbation of the resonance state[58] or for calculation of the cross section in the region of frequencies where the ionization process has a direct nature. From the data of Alimov[59] it follows that the observed resonance is of the order of 10 cm^{-1}.

At the present time there is no method which permits a quantitative description of a strong perturbation of bound electron states, which is observed in the experiments described.[14] The results of experiments on observation of resonance ionization in a strong field should indicate the specific physical phenomena which the theories must describe.

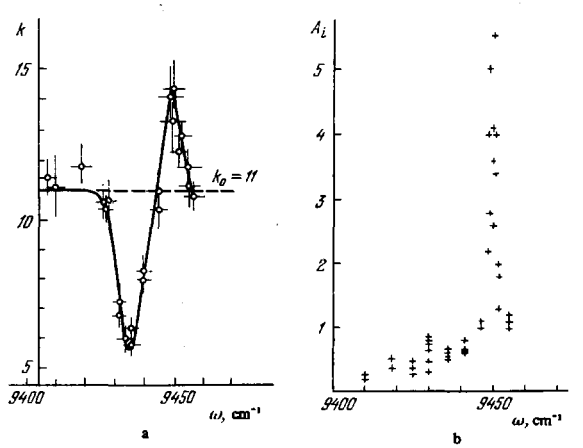

FIG. 11. Experimental data on resonance eleven-photon ionization of the Xe atom, observed at a field strength ≈5×10^7 V/cm with linear polarization of the light. a) The quantity $k = \partial \log A_i / \partial \log Q$ as a function of frequency; b) ion signal pulse height A_i as a function of frequency (Q is the energy in the laser pulse; data of refs. 43 and 44).

If we turn to experiments on ionization of atoms in a very strong field, the natural question arises as to how universal are the conclusions drawn from experiments with xenon. Although the degree of nonlinearity of different processes is significantly different (from $k_0 = 11$ for xenon to $k_0 = 22$ for helium in the case of a neodymium glass laser), the adiabaticity parameter varies weakly, lying in the range $10 \geq \gamma \geq 5$, and the field strength at which ionization is observed also does not change greatly (from 5×10^7 in the case of xenon to 10^8 in the case of helium). Most experiments with noble gas atoms have been carried out with a fixed frequency; in practically all cases the dependence of ionization probability on radiation intensity is not described by Eq. (1), and approximation of the experimental data by a power law gives a value $k < k_0$ (see for example ref. 42). The results of the experiments described above with variable frequency provide a basis for suggesting that in all experiments carried out with a fixed frequency an intermediate resonance arises[57]. The relatively large width of the radiation spectrum used in experiments with a fixed frequency (~10 cm^{-1}) and the high degree of nonlinearity of the ionization processes observed result in a large reduced width of the radiation line, which favors this suggestion. Beyond doubt, great interest is presented by the experimental verification of this idea.

c) **Main results of research.** The first conclusion permitted by the research results is that for $\gamma > 1$ the resonance ionization process is observed over the entire range of variation of the principal parameters describing the field and the atomic system. The second conclusion is that there are three qualitatively different cases: the intermediate resonance arises with a bound state practically unperturbed by the radiation field, with a weakly perturbed state where the perturbation can be considered a small correction to the unperturbed state, and with a strongly perturbed state which can be described only as a quasistationary state of the system atom + light field. The degree of perturbation of the resonance state is determined first of all by the field strength, although the distance between the levels can also be important. A third conclusion is that the resonance ionization process is closely related to various phenomena arising on perturbation of bound electron states by a light field. Only by having the possibility of qualitatively and quantitatively describing the perturbation of the resonance state can we describe the resonance ionization process. On the other hand, investigation of the resonance ionization process is necessary for description of the perturbation of bound electron states. Finally, we should mention that as a whole the studies of these interrelated phenomena are in their initial stages where each new result presents substantial interest.

CONCLUSION

In concluding this review of investigations of the multiphoton ionization of atoms, it is necessary to turn to those questions which for various reasons have not been discussed.

The first and perhaps the most important question involves the properties of the electrons produced in the

ionization. The energy, angular directivity, and polarization of the electrons have been studied theoretically in some detail.[28,29,60] However, up to the present time the properties of the electrons produced have not been investigated in sufficient detail.[15)] The absence of experimental data has been a justification for delaying discussion of this question for some time. It should be noted that the qualitative difference of the spectra of electrons produced under the action of linear and circular fields was predicted by several authors[28], both in the transition region for $\gamma \sim 1$ and in the adiabatic region for $\gamma \ll 1$. This difference, which is based on the conservation of angular momentum, is fundamental. This difference shows up only weakly in the integral characteristics of the ionization process. The question of the role of various collective effects also has not been discussed. The possibility of appearance of correlation effects in a complex atom indicates the necessity of considering them even for justification of use of the single-electron approximation. This interesting question still has not been discussed for conditions where the ionization occurs in a strong light field.

Finally, nothing has been said of the tunnel effect in a light field. Unfortunately, the experimental situation does not permit us to hope that interesting results will be obtained. This can easily be seen from Fig. 1–the condition $\gamma \ll 1$ is satisfied only for field strengths of the order of the intra-atomic field, if we remain in the optical frequency region. The condition $\gamma \ll 1$ can be achieved if the radiation frequency is substantially decreased, by going to the far infrared, perhaps using a CO_2 laser ($\lambda \sim 10\ \mu$), which permits a high field strength to be obtained. However, the great difference in frequency does not permit us, on the basis of data obtained in the visible region for $\gamma \geq 1$, to draw any quantitative conclusions as to the ionization probability and the role of saturation in the far infrared region and the practical possibilities of studying the ionization process for $\gamma \ll 1$. It may turn out that an answer to the fundamental question of the transition of the multiphoton process to tunneling in a variable field can be obtained more quickly and easily by observing the ionization of negative ions or the external surface photoeffect. The results of recent experiments[61] confirm this idea. We recall that it became clear even from the theory of L. V. Keldysh that the nature of the potential well does not have an important effect on the qualitative behavior of the ionization process in a variable field.

In order that this lengthy enumeration of uninvestigated questions not produce a false impression of the absence of progress in study of the multiphoton ionization of atoms, we will conclude by briefly formulating once again the main conclusions from experimental and theoretical studies which can now be considered firmly established.

The first and, we believe, most important conclusion which can be drawn from studies of ionization of atoms in a strong light field is that for $\gamma > 1$, depending on the light frequency, the ionization process has either a direct or resonance nature.

In regard to the direct process, the most important conclusion is that the ionization process occurring for field strengths $<10^7$ V/cm, for $\gamma > 10$ and for a significant difference between the energies $k\hbar\omega$ (where $1 \leq k \leq k_0$) and the energies of the unperturbed bound states of the electron in the atom, is satisfactorily described by nonstationary perturbation theory of the k_0-th order using as a basis the unperturbed spectrum of the atom.

The most important conclusion from studies of the resonance process is the demonstration of the possibility of obtaining information on the spectrum of quasistationary states of the electron in the atom; in just this area, data have been obtained on the constants of the quadratic perturbation of the bound electron states in a light field.

A specific feature of the multiphoton ionization process is that under the influence of a strong light field the electron not only transfers from one state to another but the states themselves are substantially perturbed by the field. Just this feature results in a close connection of the study of multiphoton ionization and the study of the spectrum of quasistationary states of the system atom + light field.

From the discussion carried out above of the ionization of atoms in a strong light field, the state of the investigations is evident–up to the present time only some general features have been determined, and the most promising directions of research have been revealed. In this situation a review of the results obtained can suffer from a certain subjectivism which can appear also in the list of references cited, which is in fact significantly more extensive.

In conclusion the author takes the occasion to express has gratitude to V. P. Kraĭnov and L. P. Rapoport, who informed him of their manuscript and who made a number of valuable remarks which were adopted in preparation of the text for publication.

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1) The results of Keldysh [1] should evidently describe the case of ionization of negative ions; unfortunately, this process has not been studied experimentally in sufficient detail. The greatest interest is presented by experiments on ionization by infrared radiation (for example, using a CO_2 laser, $\lambda \approx 10\ \mu$), since they can provide information on the ionization process for $\gamma < 1$ (see Fig. 1)

2) Getting ahead of ourselves, we note that the ionization of an atom differs from the removal of an electron from a rectangular potential well only in the pre-exponential factor; the exponential is not changed (see Chapter 2).

3) See for example the review by Bonch-Bruevich and Khodovoĭ [62].

4) See for example the book by Sobel'man [63].

5) Here and subsequently the interaction is assumed dipole, since the wavelength of the light is much greater than the size of the atom.

6) The quasienergy method is described in the review article by Zel'dovich. [64]

7) Strictly speaking, criteria of applicability of non-stationary perturbation theory must be formulated in the language of variation of wave functions (see for example ref. 65); however, from a practical point of view it is desirable to formulate criteria in terms of the observed quantities.

8) In what follows we set forth the standard approach to perturbation theory, whose basic assumptions are given at the beginning of the chapter. In principle an expansion can be made also in other functions, for example, in functions describing the free electron in the field of the wave (see ref. 66).

9) The ratio $W^{lin}_{k_0}/W^{circ}_{k_0} = (2k_0 - 1)!!/k_0! > 1$ obtained in ref. 67 is valid only for the hydrogen atom, in which there is degeneracy in orbital angular momentum.

10) Methods of using the Green's function in perturbation theory are described in the monograph by Baz' et al [68].

11) It must be recalled that the direct ionization process is described by Eq. (1), and therefore the dimensionality of the multiphoton cross section $\alpha_{k_0} = W/F^{k_0}$ is $[\alpha_{k_0}] = cm^{2k_0}\ sec^{k_0-1}$ for dimensionalities $[W] = sec^{-1}$ and $[F] = cm^{-2}\ sec^{-1}$.

[12]A discussion of the possibilities of use of the quasiclassical method for solution of the problem of ionization of an atom in a variable field has been given in the monograph by Baz' et al. [68] cited above.

[13]This question has been discussed in detail in the physics course by Landau and Lifshitz [69].

[14]A particular but important question is the solution of the problem of ionization for $I \sim \hbar\omega$ without limitations in field strength.

[15]The first experimental data on the spectrum of emitted electrons in multiphoton ionization of an atom have been reported in [70].

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