Ionization of atoms in an intense light field

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The dependence of the nature of the multiphoton ionization process in the xenon atom on the frequency of linearly and circularly polarized fields was investigated experimentally for a value of the adiabaticity parameter $\gamma \approx 5$. Depending on the frequency, a direct ionization process as well as the appearance of intermediate resonances involving bound states of an electron in an atom are observed in a linearly polarized field. The observed resonances indicate a strong perturbation of the bound-state spectrum by the external field. Resonances are not observed in a circularly polarized field.

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1. INTRODUCTION

Beginning with the work by Keldysh,[1] it is known that the ionization of an atom (characterized by an ionization potential I) in an alternating electromagnetic field (characterized by an electric field strength $\mathscr{E}$ and a frequency ω) is determined by the value of the adiabaticity parameter

$$\gamma = \frac{\omega}{\omega_{tun}} = \left(\frac{2m}{e^2}\frac{\omega^2 I}{\mathscr{E}^2}\right)^{1/2} \quad (1)$$

(ω_{tun} denotes the tunneling frequency, and m and e denote the electron's mass and charge). The ionization of atoms in a light field with $\gamma \gg 1$ (the multiphoton limiting case) has been investigated in sufficient detail.[2] Depending on the degree of polarization, the frequency, and the intensity of the light field, the ionization process at $\gamma \gg 1$ may be of a direct nature (when the transition of an electron from the ground state to the continuous spectrum may be described as a series of virtual transitions) as well as of a resonance nature (when an intermediate—as a rule, multiphoton—resonance appears involving an excited, bound electron state).

We note that if one is talking about atoms and the light band of frequencies, the case $\gamma \gg 1$ corresponds to relatively small intensity of the light field ($\mathscr{E} \lesssim 10^6$ V/cm), which is in any event small in comparison with that limiting field strength which can be attained by using contemporary lasers ($\sim 10^{10}$ V/cm). Obviously a real possibility exists to also achieve ionization of atoms in the other limiting case, when $\gamma \ll 1$, and the ionization process has the nature of a tunneling effect. However, it is not possible to investigate this case at optical frequencies. The point is that the ionization probability already becomes so large for $\gamma \sim 1$ that, even for ultrashort, picosecond laser pulses, the efficiency of ionization during the pulse reaches a value of the order of unity.

Investigations of the ionization process in the intermediate case, when $\gamma \sim 1$, are of considerable interest if only because such a situation is realized in the optical band for atoms having large ionization potentials (for example, noble gas atoms). Many experimental articles have been devoted to the investigation of ionization for $\gamma \sim 1$[2]; however, the only important result of these investigations consists of the observation of intermediate resonances.[3] The theoretical description of ionization for $\gamma \sim 1$ is hindered both from the point of view of the fulfillment of the criteria for the applicability of the various methods (this is the region where, strictly speaking, neither the standard approach of perturbation theory nor the adiabatic (quasiclassical) approximation is applicable), and because one should anticipate[2] that the spectrum of the system "atom + field" differs substantially from the unperturbed spectrum. In actual fact, for $\gamma \sim 1$ ionization is observed in the presence of strong fields, whose field strengths lie in the interval $10^{-2}\mathscr{E}_{at} \leq \mathscr{E} \leq 10^{-1}\mathscr{E}_{at}$ (see[2]).

The present article is devoted to an extension of our investigations[3] of the ionization of atoms in a light field for a value of the adiabaticity parameter $\gamma \sim 1$. The region of investigated frequencies was substantially enlarged in order to obtain new information in comparison with previous experiments, but the main feature is that measurements were carried out for different degrees of elliptical polarization of the light. The experiments were performed on the xenon atom.

2. THE EXPERIMENTAL SETUP

In its main features the experimental procedure was standard for investigations of the multiphoton ionization process in atoms (see, for example,[2]). Therefore, we mention only certain details which are unique for the present measurements. The intense light field was created by focusing the radiation from a powerful Q-switched laser neodymium-glass ($\hbar\omega \approx 1.17$ eV). A rotating prism was used as the Q-switch. A dispersion resonator was used to vary the generation frequency. The dispersion element was a polarization-interference Wood's filter,[4] which consists of two polarizers (Glan–Foucault prism) with an anisotropic quartz plate placed between them; the plate temperature could be varied and when necessary maintained constant with the required accuracy. The Wood's filter ensured purely linear polarization of the generated light. By placing a diaphragm in the resonator, only axial transverse modes occurred in the regime of generation. In order to increase the radiation intensity, two amplifying stages operating in the traveling-wave regime were utilized. A quarter-wave plate of crystalline quartz was placed in the beam of light from the laser at the output from the second amplifier. Variation of the orientation and temperature

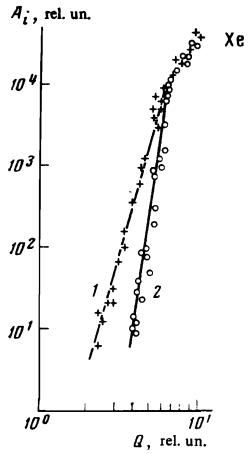

FIG. 1. Typical result of measuring the dependence of the amplitude A_i (in relative units) of the ionic signal on the radiation energy Q (in relative units) for two different radiation frequencies: 1) $K=5.77\pm0.5$, $\omega=9432$ cm^{-1}; 2) $K=10.85\pm0.47$, $\omega=9424.5$ cm^{-1}. An approximate plot of the power law $A_i\sim Q^K$ is drawn in the region where there is no saturation of the ionic signal.

of this plate allowed us to obtain light with the required degree of elliptical polarization from the initial linearly polarized light.

The laser radiation was focused in a vacuum chamber, filled to a pressure $\approx10^{-4}$ Torr with the gas under investigation. Under such conditions no collisions of electrons with atoms arose in the region of light focusing ($\sim10^{-2}$ cm) during the duration of the laser pulse ($\sim10^{-8}$ sec). The ions, which were produced in the light-focusing region, were accelerated by the constant field ($\sim10^2$ to 10^3 V/cm), were focused by the electronic-optical system, and were detected by an electron multiplier. Measurement of the intensity of the radiation in the region of ion production was carried out by the standard method, by an independent measurement of the energy, and of the temporal and spatial distribution of the laser radiation.[2]

The measured quantities were: the functional dependence of the ionization probability W on the intensity F of the radiation for a series of fixed light frequencies; the dependence of the ionization probability on the frequency for a fixed intensity; the dependence of the ionization probability on the elliptical polarization of the light for a fixed frequency and intensity of the radiation. The necessary remarks concerning the procedure for measuring the various quantities are cited in the appropriate parts of the following section.

In conclusion we note that since the dependence $W(F)$ is very strong in the case under investigation (it is approximately described by the relation $W\sim F^{11}$), the dynamical range of the apparatus amounted to $\sim10^3$ times, and the maximum value of a detectable probability was always of the order $W=\tau^{-1}\approx10^8$ sec^{-1}; then practically all measurements are carried out at a field strength $\mathscr{E}\approx5\times10^7$ V/cm and for a value of the adiabaticity parameter $\gamma\approx5$.

3. RESULTS OF THE MEASUREMENTS AND THEIR DISCUSSION

1. Linear polarization of the light

a. $W(F)$ as a function of the frequency of the radiation. The measurement procedure consists of the following. A definite frequency of laser generation was fixed, and the operating regime of the laser was carefully stabilized with respect to the amount of energy in a pulse; the energy passing through the chamber was varied in a series of successive pulses of the laser; the variation of the radiation energy was achieved by linear attenuation at the entrance to the vacuum chamber due to the reflection of part of the light from the glass plates; the amplitude A_i of the ion signal and the energy Q of the radiation were measured. For a constant space-time distribution of the radiation in the region of ion production, the $A_i(Q)$ dependence is equivalent to the desired $W(F)$ dependence. The dispersion in the amplitude A_i of the ion signal associated with a fixed Q is a measure of the invariance of the space-time distribution. All measurements were carried out under conditions when it would be possible to observe the region of saturation of the ion signal, where

$$\int_0^\tau W\,dt\sim1.$$

and thereby reliably isolate that region where

$$\int_0^\tau W\,dt\ll1.$$

In the second region of interest to us, the set of individual values $A_i(Q)$ was approximated by the power-law relationship $A_i\sim Q^K$. The quantity $K=\partial\log A_i/\partial\log Q$ was the final result of the measurement. The results of two typical measurements are shown in Fig. 1. The sum of the results of all individual measurements is given in Fig. 2 in the form of a plot of $K(\omega)$. We note that the amplitude of the variation in the quantity K exceeds the accuracy of the measurement ΔK by approximately an order of magnitude.

It is quite clear from Fig. 2 that, depending on the frequency of the light, values of K are observed which are equal to the value K_0, where

$$K_0=\langle\tilde{I}/\hbar\omega+1\rangle, \tag{2}$$

as well as values of K which are smaller or larger than K_0. (Here $\tilde{I}=I+E_{osc}$ is the effective binding energy of an

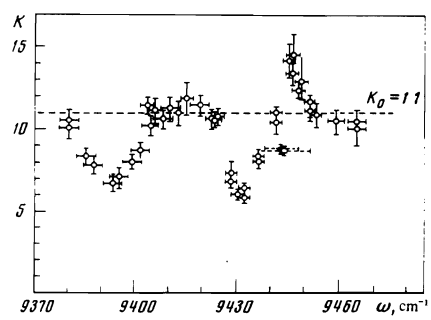

FIG. 2. Dependence of the quantity $K=\partial\log A_i/\partial\log Q\sim\partial\log W/\partial\log F$ on the frequency of the linearly polarized light. ○—results of the present experiment; ■ and □—results of previous experiments performed in laboratories at Saclay (France) and in FIAN for a fixed frequency of laser generation.[2]

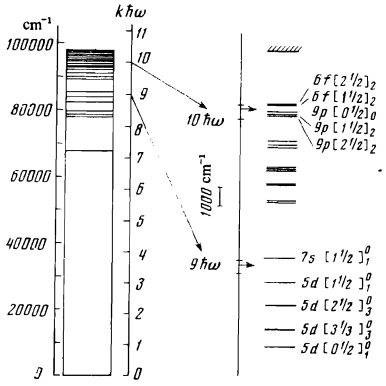

FIG. 3. The bound-state spectrum of the xenon atom. To the right in magnified scale—the region of localization of the unperturbed bound electron states, the transition energy to which is of order $9\hbar\omega$ and $10\hbar\omega$. The ranges of variation of the transition energy, realized in the experiment, are indicated. The arrows indicate the value of the energy corresponding to a resonance frequency $\hbar\omega \approx 9440$ cm^{-1}.

electron in a strong alternating field; $E_{osc} = e^2\mathscr{E}^2/4m\omega^2$ is the oscillating energy of a free electron in the field of the wave, equal to ~0.5 eV for $\mathscr{E} \approx 5\times10^7$ V/cm; $\langle X\rangle$ denotes the integer part of the value X). A similar dependence $K(\omega)$ was also observed earlier in investigations of the ionization process for $\gamma \gg 1$, where the deviations of K from K_0 were uniquely identified as the effect of an intermediate resonance with a bound excited electron state.[2,6,7] Therefore, one can already confirm from one form of the $K(\omega)$ dependence that the ionization process is of a direct nature at a frequency 9405 cm$^{-1} \leq \omega \leq$ 9425 cm^{-1}, but intermediate resonances involving bound electron states appear at frequencies 9380 cm$^{-1} \leq \omega \leq$ 9405 cm^{-1} and 9425 cm$^{-1} \leq \omega \leq$ 9465 cm^{-1}.

In order to obtain quantitative information characterizing the intermediate resonances, it is necessary to take the bound state spectrum of the xenon atom into consideration. The nature of this spectrum, in which the energy of the first excited state is of the order of 8 eV, indicates that a resonance may arise only upon the absorption of 9 or 10 quanta (Fig. 3). Precisely in such a case, when the degree of nonlinearity of the transition from the ground state o to the resonant state r is substantially greater than the degree of nonlinearity of the transition from the resonant state r to the continuous spectrum ($K_{or} > K_{rE}$), the frequency dependence of the ionization probability is described[5]1) by Keldysh's resonance formula[1]:

$$W_{oE}^{(K_o)} \approx \frac{|V_{or}^{(K)}|^2}{[E_r(\mathscr{E}) - E_o(\mathscr{E}) - K\hbar\omega]^2 + [\gamma_r(\mathscr{E})]^2} W_{rE}^{(K_o - K)}. \qquad (3)$$

In a comparison of the experimental data with expression (3), it is always necessary to keep in mind instrumental effects which broaden the resonance—the Doppler effect, the finite linewidth of the laser radiation, and inhomogeneous broadening associated with a nonuniform distribution of the radiation intensity over the target.[2,6] Quantitative estimates of the instrumental effects showed that in the present case they are small in comparison with the experimentally observed width of the resonances; therefore, we shall not take them into consideration below.

A quantitative comparison of relationship (3) with the experimental data without additional assumptions or calculations can be carried out, when $K_0 - K = 1$, by retaining only the first (quadratic) term in the expansion of the dynamic polarizability with respect to the field (E_r, E_0, $\gamma_r \sim \mathscr{E}^2$) and introducing the dimensionless parameter $\eta = \gamma/\Delta$, where $\Delta = |E_r(\mathscr{E}) - E_0(\mathscr{E}) - K\hbar\omega|$ denotes the resonance detuning. Numerical calculations show that the $K(\omega)$ dependence is well described for $\eta = \frac{1}{4}$ in the frequency range $\omega > 9425$ cm^{-1} (Fig. 4). Since the scale of the interval of frequency variation at which the resonance appears is ~10×(9465 – 9425) cm^{-1} ≈ 400 cm^{-1}, one must assume that the variation of the transition energy from the ground state to the resonance is ~400 cm^{-1}, and the width of the resonance is ~100 cm^{-1}.

Exact resonance ($\Delta = 0$) is achieved at a frequency 9444 ± 3 cm^{-1}, which corresponds to a transition energy E_{or} equal to 94440 ± 30 cm^{-1} in a field of electric field strength $\mathscr{E} \approx 5\times10^7$ V/cm (for $K_0 = 11$ the condition $K_0 - K = 1$ corresponds to a ten-photon resonance).

For $\omega < 9405$ cm^{-1} the $K(\omega)$ dependence can not be described within the scope of the assumptions made above. More accurately the data measured in the frequency interval from 9380 to 9405 cm^{-1} represent only part of the resonance dependence of $K(\omega)$, with the other part being observed at frequencies $\omega < 9380$ cm^{-1}.

Before reaching any conclusions based on the results of an analysis of the $K(\omega)$ dependence, let us turn to an experimental investigation of the frequency dependence of the ionization probability.

b. The $A_i(\omega)$ dependence. Measurement of the frequency dependence of the ionic signal A_i (a quantity proportional to the ionization probability) for a fixed intensity of radiation is the most accurate method of investigating intermediate resonances.[2] However, in the case of interest to us the potentialities of this method are restricted by the high field strength and the highly nonlinear nature of the ionization process. The high field strength causes a strong perturbation of the resonance state and a large attenuation of this state and thereby causes a small difference between the ionization probability at exact resonance and upon large detuning. The large degree of nonlinearity causes a large amount of scatter in the ionic signal at a fixed energy in the radiation pulse, due to fluctuations in the space-time

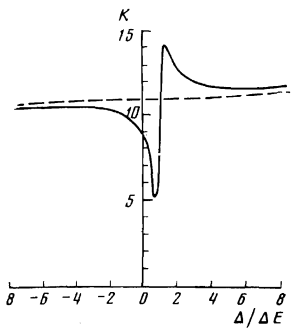

FIG. 4. Numerical calculation according to formula (3) of the dependence of the quantity K on the frequency of the radiation. Δ denotes the initial detuning off resonance, and ΔE denotes the shift of the resonance state. The calculation is carried out for the value of the parameter $\eta = \gamma/\Delta = \frac{1}{4}$.

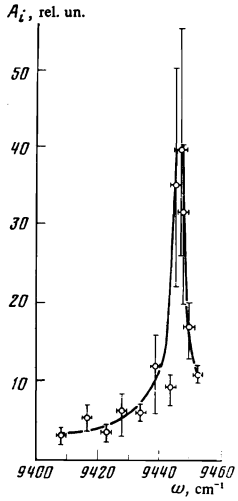

FIG. 5. Dependence of the amplitude A_i of the ionic signal (in relative units) on the radiation frequency ω at a fixed value of the radiation intensity.

distribution of the radiation. However, good stabilization of the operating regime of the laser enabled us to measure the $A_i(\omega)$ dependence in the frequency interval from 9410 to 9455 cm^{-1} (Fig. 5). From these data it is evident that the $A_i(\omega)$ indicates the existence of a resonance at the frequency 9447 ± 1 cm^{-1}, which coincides within the limits of experimental accuracy with the data following from measurements of the $K(\omega)$ dependence. The half-width of the resonance maximum in the ion output has a value 7 ± 1 cm^{-1}. If it is assumed that the resonance arises upon the absorption of ten quanta, then the half-width of the resonant state in the atom spectrum has a value 70 ± 10 cm^{-1}; the result which follows from measurements of the $K(\omega)$ dependence also agrees satisfactorily with this value.

c. Discussion of the data on the resonance. Summarizing the results of the measurements of the dependences $K(\omega)$ and $A_i(\omega)$ in the frequency range $\omega > 9425$ cm^{-1}, one can state that an intermediate resonance is observed upon the absorption of ten quanta, and in a field of electric field strength 5×10^7 V/cm this resonance is quantitatively characterized by three quantities: the transition energy $E_{or}(\mathscr{E}) = 94470 \pm 10$ cm^{-1} from the ground state to the resonance; the half-width $\gamma_r(\mathscr{E}) = 70 \pm 10$ cm^{-1} of the resonance state; the change $\delta E_{or}(\mathscr{E}) \sim 400$ cm^{-1} in the transition energy from the ground state to the resonance state under the influence of the field.

If we return to the spectrum of the xenon atom in the absence of a field, a transition energy $E \approx 94000$ to 95000 cm^{-1} corresponds to the region of localization of the $9p$, $8d$, and $6f$ multiplets in the series of levels to which a ten-photon transition is allowed from the ground state. The average distance between the levels in the $9p$, $8d$, and $6f$ multiplets is of order $\Delta E_{ij} \approx 10^2$ cm^{-1}. By comparing this value with those values which characterize the experimentally observed perturbation of the atomic spectrum, it is clear that $\delta E_{or}(\mathscr{E})$, $\gamma_r(\mathscr{E}) \gtrsim \Delta E_{ij}$, i.e., the perturbation of the spectrum is strong. From our point of view this experimental fact indicates that a theoretical analysis of such resonances cannot be based on data concerning the unperturbed atomic spectrum, and also on an extrapolation of data measured in weak fields, and cannot be carried out within the framework of perturbation theory.

The problem of the construction of quasistationary states of the "atom+field" system by intermixing the initial unperturbed states arises in such a situation. The theoretical solution of such a problem is greatly facilitated for a finite number of initial states. A similar calculation was carried out[8] on the basis of an assumption about predominant intermixing of the various states of the $9p$ and $7s$ multiplets, the frequency of the transitions between these states being close to the frequency of the external field. The methods, previously applied to a calculation of intramultiplet intermixing,[9] were utilized. The calculation[8] did not give even qualitative agreement with the experimental data on the resonance. From our point of view, this discrepancy may serve as an indication of an unnecessary simplification, which was made in the model adopted in[8]. Thus, for example, the absence in[8] of a self-consistent calculation of the field broadening of the intermixed states is, in fact, not justified since the transitions $7s \rightarrow 9p$, just like transitions from the $9p$ multiplet to the continuum, are of one-photon character.

In spite of the assertions made above concerning the inapplicability of perturbation theory and of the weak-field experimental data, to an analysis of the situation which arises in a strong field, one can make one interesting comparison. We have in mind the width of the excited atomic state, resulting from a one-photon transition to the continuous spectrum. The entire set of experimental and theoretical data on photoionization, when extrapolated to an intensity $F \approx 10^{32}$ photons $\cdot$ cm^{-2} $\cdot$ sec^{-1} (which corresponds to an electric field strength $\approx 5 \times 10^7$ V/cm) gives a level width of the order of several thousand cm^{-1} (see, for example,[10]). This estimate exceeds the experimentally measured width by more than an order of magnitude. We note that from the point of view of perturbation theory, the width can only be larger due to the neglect of higher order terms. The relatively small width observed experimentally may be caused by the tunneling nature of the transition in the presence of a strong field. Qualitatively the reduction of the ionization probability for $\gamma \sim 1$ in comparison with the extrapolation of the data according to perturbation theory was already evident from Keldysh's adiabatic theory.[11] This same result follows from the exact solution of the problem of the extraction of an electron from a short-range potential in a circularly polarized field.[11]

2. Circular polarization of light: the dependence $K(\omega)$ as a function of the frequency of the radiation

The measurement procedure[12] was completely analogous to the procedure used for linear polarization of the radiation (see Sec. 3, part 1*a*). The set of results of the individual measurements is presented in Fig. 6 in the form of the $K(\omega)$ plot. The sharp difference in the nature of this dependence from $K(\omega)$ dependence for linear polarization of the light (Fig. 2) is clear. In the case of circular polarization in that same range

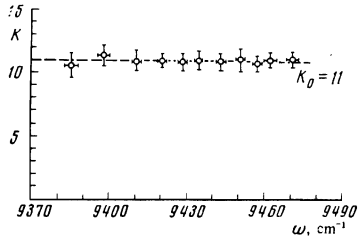

FIG. 6. The value of K as a function of the frequency of circularly polarized radiation.

of variation of the light frequency, the value of $K(\omega)$ is practically constant and is given by $K_0 = 11$ within the limits of accuracy of the measurements.

Thus, resonances are not observed in the interval of frequency variation from 9380 to 9470 cm^{-1} for circular polarization of the radiation. Making the assumption already discussed above concerning the possible appearance of a reasonance associated with the absorption of ten photons, one can state that resonances do not appear in the region of the atomic spectrum having a value ~1000 cm^{-1}. In our view the absence of resonances for circular polarization of the light is related to the specific characteristics of a circularly polarized field and to the selection rules for multiphoton transitions. If one starts from the unperturbed atomic spectrum, one can see that the levels to which nine- or ten-photon transitions (levels having orbital angular momentum $l = 9$, 10) are allowed, are located in the immediate vicinity of the ionization limit and have energies which exceed the energy $10\hbar\omega$ by a thousand cm^{-1}.

4. CONCLUSION

The entire collection of experimental facts enables one to reach three fundamental conclusions about the ionization of atoms when the value of the adiabaticity parameter $\gamma \sim 1$: 1) the nature of the ionization process is essentially determined by the polarization of the light; 2) depending on the frequency and polarization of the light, the process may be either direct in nature or have a resonance character; 3) the spectrum of the bound electron states is strongly perturbed by the light field. It is necessary to note that all of these conclusions are qualitatively confirmed by the experimental data obtained in connection with the ionization of the xenon atom.[3] One would anticipate that the nature of the ionization process would be the same for all noble gas atoms since, for example, for infrared light with $\lambda \sim 1\mu$, the field strength at which ionization of all atoms is observed differs in all by a factor of 2 or 3, but the value of γ is essentially the same and amounts to several times unity.

The observation of the direct ionization process for linear and circular polarization of light first revealed the possibility of measuring the absolute value of the ionization probability and the dependence of the probability on the degree of elliptical polarization of the light and the comparison of these quantities with calculations carried out by using perturbation theory and the quasiclassical method.

Finally, let us turn our attention to the absolute value of the degree of nonlinearity of the direct ionization process, observed for $\gamma \approx 5$ in the experiments described above. At first glance the experimental result ($K \approx 11$) contradicts the conditions of its execution ($\gamma \approx 5$) since according to Keldysh's theory[1] the degree of nonlinearity, given by $K_0 = 11$, should be observed only for $\gamma \gg 1$. The exact calculation,[13] carried out for circular polarization of the radiation within the framework of the quasiclassical (adiabatic) approximation gives a dependence $W(F)$ for $\gamma \approx 5$ which is different from a power-law dependence. However, quantitatively this difference is small and upon approximation of $W(F)$ by a power law over the narrow range of variation of the quantity F which was achieved in our experiments, this calculation gives $K = 10.6$. The accuracy of the experiment $\Delta K \approx 0.7$ does not allow us to distinguish this value from $K_0 = 11$.

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[1)]Also see the article by Kazakov *et al.* on p. 20 of this issue.

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