Multiphoton Resonance Ionization of Atoms

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The conditions which are optimal for observation of multiphoton resonance with a bound electron state during multiphoton ionization of an atom are considered. An experiment is described in which three-photon resonance with the 4f level is observed in four-photon ionization of the potassium atom. The experiments show that the presence of resonance significantly alters the multiphoton ionization probability as well as the functional dependence of the probability on the radiation field strength. An analysis of the experimental results shows that the ionization process occurs in two steps, the first being three-photon excitation of the atom to the 4f state; this is then followed by one-photon resonance excitation of the excited atom. Resonant excitation of the 4f state depends significantly on the Stark shift of the state in the radiation field. The cross section for three-photon excitation is estimated. The result is in good agreement with perturbation-theory calculations.

1. INTRODUCTION

WHEN an atom is ionized in a strong electromagnetic field of optical frequency, a situation may arise wherein the energy of several quanta turns out to be equal to the energy of the bound state of the electron. In this case the ionization will proceed in two stages, multiphoton resonant excitation of the atom and ionization of the excited atom. The second stage can also have the character of a multiphoton transition. A distinguishing feature of the conditions for the observation of multiphoton ionization is a large electric-field intensity. The action of a strong field leads not only to the extraction of the electron from the atom, but also to an appreciable perturbation of the bound states of the electron.

The first theoretical analysis of the multiphoton ionization of an atom in the presence of resonance is contained in the well-known paper of Keldysh,^[1] where the principal effect, the perturbation of the resonant level by the radiation field, was taken into account. The results obtained by Keldysh show that the occurrence of resonance can lead to a change in the dependence of the ionization probability W on the radiation intensity F compared with the well-known relation

$$W = \alpha F^{k_0}, \quad k_0 = \langle I/\hbar\omega + 1 \rangle, \tag{1}$$

where k_0 is the number of quanta absorbed during the ionization. Several theoretical papers^[2-7] are devoted to different particular questions concerning multiphoton ionization in the presence of resonance.

In the overwhelming majority of experiments devoted to the investigation of multiphoton ionization of atoms one observes a power-law dependence of the ionization probability on the radiation intensity, $W = \alpha F^k$, with an exponent $k < k_0 = \langle I/\hbar\omega + 1 \rangle$.^[8] This result may be due, in particular, to the occurrence of resonances. However, the large intensity of the radiation field at which multiphoton ionization is observed, and accordingly the strong perturbation of the atomic spectrum, as well as the impossibility of taking this perturbation accurately into account, do not make it possible to interpret these results with sufficient accuracy. On the other hand, our experiments on four-photon ionization of the sodium atom by radiation from a neodymium-glass laser have shown that in these cases the exponent is $k = k_0$.^[9] These results were

subsequently confirmed by Agostini et al.^[10] From an analysis of the experiments^[9,10] it follows that the difference between the energy $k < k_0$ of the quanta and the energy of the bound states of the electron is large in these cases, and that the nearest levels are weakly perturbed in the field of intensity $\sim 10^6$ V/cm in which the ionization was observed, so that the energy differences do not change significantly. We have assumed that the conditions most favorable for the investigation of resonant multiphoton ionization of atoms are precisely those in which the levels are weakly perturbed, so that resonance with an individual isolated level can be realized. We performed the experiment aimed at observing the resonance^[11] on the potassium atom, using a neodymium-glass laser with variable generation frequency. By varying the frequency, we were able to produce resonance between the energy of three quanta and the energy of the 4f level. The present paper is devoted to this experiment.

2. THEORETICAL ANALYSIS OF THE MULTIPHOTON IONIZATION PROCESS IN THE PRESENCE OF RESONANCE

Keldysh^[1] has considered an atomic model having one level s, the transition to which from the ground state has the character of a multiphoton resonant transition. The probability $W_{0E}^{(k_0)}$ of k_0 -photon ionization of the atom in the presence of k-photon resonance with the level s is described by the relation^[1]

$$- \frac{W_{\delta E}^{(k_0)} = W_{\delta E}^{(k_0)} W_{\delta E}^{(k_0-k_0)}}{(E_0 - E_s - k\hbar\omega + \Delta E_s(F))^2 + \hbar^2 (W_{\delta E}^{(k-k_0)} + \Gamma_s)^2} \beta_{s E} F^{k_0-k},$$
(2)

where $W_{0S}^{(k)}$ is the probability of k-photon resonant excitation of the atom to the state s, $W_{SE}^{(k_0-k)}$ is the probability of ionization from the excited state s; E_S , $\Delta E_S(F)$, $W_{SE}^{(k_0-k)}$ and Γ_S are the energy, Stark shift, broadening, and proper width of the resonant level s, and $\Delta_0 = E_0$ $- E - k\hbar\omega + \Delta E_S(F)$ is the deviation from resonance. Formula (2) enables us to analyze qualitatively the ionization process in the presence of resonance (see Fig. 1). In a weak field, when the condition

$$\Delta E_s(F), W_{sE}^{(k_0-k)} < \Gamma_s, \tag{3}$$

is satisfied, i.e., when the Stark shift and the broadening of the resonant level are smaller than its proper width, the corresponding term in the resonant denominator (2) can be neglected and the probability of the kphoton excitation is proportional to the radiation intensity raised to the power k, $W_{oS}^{(k)} \sim F^k$. If the transition from the excited state s to the continuous spectrum is not saturated, then $W_{SF}^{(k_0-k)} \sim F^{k_0-k}$. The k₀-photon ionization process is described in this case by the relation (1), $W_{oE}^{(k_0)} \sim F^{k_0}$.

When the field intensity increases, both the ionization probability and the functional dependence of the probability on the radiation intensity change. On the one hand, owing to the increase of the probability of the transition from the state s into the continuous spectrum, saturation can set in with respect to this transition. The condition for the occurrence of saturation is the relation $W_{sE}^{(k_0-k)}\tau \sim 1$, where τ is the time during which the radiation acts on the atom (the duration of the laser-radia-tion pulse).¹⁾ When the second stage of transition saturates, the ionization process is determined in practice only by the first stage, the process of multiphoton resonant excitation of the atom into the state s, i.e., in particular, the $W_{0E}^{(k_0)}(F)$ dependence will not have the form $W \sim F^k$. So long as the condition (3) is satisfied, the $W_{0E}^{(k_0)}(F)$ dependence will be of the form $W_{0E}^{(k_0)} \sim W_{0S}^{(k)}$ $\sim F^k$. On the other hand, when the Stark shift and the broadening of the resonant level become larger than its natural width, it is impossible to neglect in the denominator of (2) the terms $\Delta E_{\rm S}(F)$ and $W_{\rm SE}^{(k_0-k)}(F),$ and the functional dependence of the probability of the resonant excitation on the radiation intensity is not described by the relation $W_{oS}^{(k)} \sim F^k$. What we have here physically is that the Stark shift of the level s upsets the resonance and the probability of the resonant excitation increases more slowly than $\sim F^k$ with increasing radiation intensity.

In the limiting case when the deviation from resonance under the influence of the field becomes very large, the resonance is lost completely, the ionization process becomes nonresonant, and the dependence of the ionization probability on the radiation intensity again takes the form (1), namely $W_{0E}^{(k_0)} \sim F^{k_0}$. Formula (2) was obtained for ideally monochromatic

Formula (2) was obtained for ideally monochromatic radiation. At a finite width of the emission spectrum Γ , there are always qualitatively the same effects. For a Gaussian character of the distribution of the radiation in the laser emission line, the probability of resonant excitation is determined by the relation^[6]

$$W_{0s}^{(h)} \sim \Gamma_{\text{eff}}^{-1} \exp\left(-\Delta^2/\Gamma_{\text{eff}}^2\right), \tag{4}$$

$$\Gamma_{\text{eff}} = [W_{sE}^{(k_0-k)}(F) + \Gamma_{k\hbar\omega}^2]^{1/2} = [W_{sE}^{(k_0-k)}(F) + k\Gamma^2]^{1/2}.$$
(5)

The foregoing relations enable us to analyze qualitatively the process of multiphoton ionization in the pres-

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FIG. 1. Calculation of the exponent of the power-law dependence of the ionization probability on the radiation intensity in accord with formula (2); Δ_0 -deviation from resonance; $\Delta E_s(F)$ -Stark shift of the resonant level; the continuous curve is drawn under the condition $W_{sE}^{(k_0-k)}/\Delta E_s(F) \approx 10^{-2}$; the dashed curve is drawn under the condition $W_{sE}^{(k_0-k)}/\Delta E_s(F) \approx 10^2$.

ence of resonance. For a quantitative analysis it is necessary to be able to calculate for a concrete level in a concrete atom, at a fixed frequency and radiation field intensity, the probability of multiphoton excitation, the probability of ionization of the excited atom, and the shift and broadening of the resonant level. In those cases when, in principle, one can employ perturbation theory for such calculations, the necessary result can be obtained by numerical calculation with electronic computers. The calculation methods used in ^[6,7] yield all the required quantities. In these papers, in particular, they calculated the experimental data obtained by us for the potassium atom, namely, the probability of four-photon ionization at large deviation from resonance, the probability of three-photon resonant excitation of the 4f level, and the Stark shift and broadening of the 4f level. Finally, it must be emphasized once more that the above approach to the description of the multiphoton ionization process in the presence of resonance is valid only if the resonant level can be regarded as isolated. i.e., so long as the change of energy or broadening of a resonant level and the levels closest to it are much smaller than the distance between levels.

3. ORGANIZATION OF EXPERIMENT

We studied the resonant multiphoton ionization of the atom by observing the four-photon ionization of the potassium atom (I = 4.34 eV) by radiation from a neodymium glass laser ($\hbar\omega \approx 1.17 \text{ eV}$) with variable generation

E, cm

1.10

K atom

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FIG. 2. Diagram of two-step four-photon ionization of the potassium atom in the presence of an intermediate three-photon resonance 2.10° with the level 4f.



 $^{^{1)}}It$ is assumed that the condition $W_{sE}\!\propto\!\Gamma_{s}~$ is certainly satisfied in this experiment.



FIG. 3. Experimental setup: 1-laser, 2-focusing objective, 3-objective for the emergence of the light from the vacuum chamber, 4 and 14-calorimeters, 5 and 6-electron-optical system for ion acceleration, 7-Faraday cylinder, 8-ion detector (electron multiplier); 9 and 13-light-splitting wedges; 10-objective similar to the working objective 3; 11-microscope, 12-coaxial photocell, 15-laser-radiation attenuator, 16-spectrograph.

frequency. In the case when the frequency of the generated radiation corresponds to the maximum of the luminescence line, the difference between the energy of two quanta and the nearest levels is on the order of several thousand cm^{-1} , and the difference between the energy of the three quanta and the nearest level 4f is 190 cm (see Fig. 2). By varying the laser generation frequency. it is possible to produce resonance between the energy of three quanta and this state. The experiment consisted of measuring the dependence of the ionization probability on the radiation intensity at different deviations from resonance. Our preceding experiments on four-photon ionization of the potassium atom by neodymium-laser radiation have shown that the ionization is observed at a field intensity $E \sim 10^6$ V/cm. Calculations performed in ^[6,7] gave for the change of the energy of the 4f level as a result of the Stark shift of this level and the ground state 4s the relation $\Delta E_{4S-4f} \approx 2.5 [10^{-6} E V/cm]^2$, and for the broadening of the 4f level the relation W_{4f-E} $\approx 0.1 [10^{-6} \text{E V/cm}]$. Consequently, at $\text{E} \sim 10^{-6} \text{V/cm}$ we have $\Delta E \approx 2.5$ cm⁻¹, and W ~ 0.1 cm⁻¹, i.e., the perturbation of the 4f level is small and in the presence of resonance one can consider the ionization process as a transition of an electron through an isolated atomic level.

From the width given above for the 4f level in the radiation field and at the laser pulse duration $\tau \sim 10^{-8}$ sec realized in our experiments it follows that the transition of the electron from the state 4f into the continuous spectrum will be saturated at $E > 5 \times 10^4$ V/cm. Thus, at $E \sim 10^6$ V/cm, in the presence of exact resonance, the process of four-photon ionization of the potassium atom should be determined by the process of



FIG. 4. Diagram of neodymium laser with variable generation frequency: 1–Q-switch (rotating prism); 2–active element; 3, 4–Fabry-Perot interferometers; 5–exit mirror; 6–diffraction-grating spectrograph.

three-photon excitation of the atom into the state 4f. A diagram of the experiment appears in Fig. 3.

The variable-frequency neodymium laser had a compound resonator (Fig. 4). The Q switch was a rotating prism. The generation frequency was varied with two Fabry-Perot interferometers placed in the resonator between the active element and the output mirror and operating in transmission. By varying the angle of inclination of the interferometer relative to the resonator axis it was possible to vary the generation frequency from the maximum of the luminescence line towards longer wavelenghts, by an amount $\approx 100 \text{ cm}^{-1}$. The generated radiation has a Gaussian intensity distribution with half-width $\approx 3 \text{ cm}^{-1}$ (Fig. 5). The spectrum of the generated radiation was measured with a diffractiongrating spectrometer. The spectral measurements were made absolute with the aid of the lines of a neon lamp. The laser operated automatically, emitting every minute an energy of several tenths of a joule, maintained accurate to several percent over hundreds of pulses. The measurement of the radiation intensity in the region where the ions were produced was carried out by standard method developed by us earlier.^[12] The energy passing through the region of ion production in the laser radiation pulse was measured with a calorimeter. To measure the space-time distribution, part of the radiation was diverted with a wedge to an objective analogous to that used to focus the main radiation flux and located at the same distance from the laser, and to a high-speed coaxial photocell of the FEK type. The spatial distribution of the radiation was measured in the focal region of the auxiliary objective. The temporal distribution of the radiation, integrated over the beam cross section, was measured with a photocell. The possibility of independently measuring the spatial and temporal distributions was checked experimentally.^[12] Control experiments have shown that the space-time distribution of the radiation does not change significantly when the generation frequency is varied. The absolute radiation intensity measurement accuracy was $\approx 50\%$.

The laser radiation was focused in the vacuum chamber onto the center of a beam of potassium atoms



FIG. 5. Dependence of the laser generation frequency on the angle of rotation of a Fabry-Perot interferometer with base $\approx 200\mu$. The generation line half-width is $\approx 3 \text{ cm}^{-1}$



FIG. 6. Arrangement of source in the vacuum chamber: 1-source, 2-housing cooled with running water; 3, 4-diaphragm, 5-atomic-beam condenser cooled with running water; 6-neodymium-laser radiation; 7-focusing region.

(Fig. 6). The atomic beam of density $n_{a}\approx 10^{12}~\text{cm}^{-3}$ was shaped with the aid of a multichannel collimator. At the indicated density in the beam, the pressure in the chamber was of the order of several units of 10^{-6} mm Hg. The ions produced in the region of intersection of the light and atomic beams were accelerated by a constant electric field, shaped by an ion-optical system, mass-divided in a mass spectrometer by time of flight, and registered with an electron multiplier. We measured the dependence of the ion-signal amplitude on the energy passing through the ion-formation region in the laser radiation pulse. The energy passing through the chamber was varied by attenuating the radiation at the input to the chamber with filters of colored glass. As is well known,^[12] at a fixed space-time distribution of the radiation, the dependence of the ion signal on the energy passing through the target is equivalent to the sought dependence of the ionization probability on the radiation intensity.

Owing to the inhomogeneity of the distribution of the radiation intensity in the region where the ions are produced, the ionization probability is different at each point of this region. One measures experimentally the summary number of ions produced in the entire volume:

$$N_i = n_a \iint W \, dv \, d\tau, \tag{6}$$

where n_a is the density of the neutral atoms. In our earlier experiments,^[8] where we investigated processes connected with the absorption of many photons in a single act, we related the total ion yield to the maximum intensity of the radiation attained in each concrete experiment. In the case of processes with a relatively small number of photons, the maximum number of ions is produced in those regions where the intensity is lower than maximal. The effective intensity, i.e., the intensity corresponding to the maximum of the produced ions, was calculated under the assumption that the probability of the ionization is connected with the radiation intensity by a power law (W ~ F^k), starting from the experimental data on the spatial distribution of the radiation.^[13] As seen from Fig. 7, which shows the result of the calculations for k = 4 and k = 10, we have F_{eff} $\approx 0.6 F_{max}$ for k = 4. The difference between the maximum and effective intensities was taken into consideration in those cases when the absolute value of the radiation intensity was significant, for example in the estiFIG. 7. Distribution of number of produced ions over regions with different intensity for four-photon (k = 4) and ten-photon (k = 10)ionization. The calculation is based on the experimentally measured distribution of the radiation intensity in the focal region.



mate of the Stark shift of the resonant level.

4. EXPERIMENTAL RESULTS AND DISCUSSION

The experimental results are shown in Figs. 8 and 9. Figure 8 shows the dependence of the ion signal amplitude Ni on the energy Q passing through the region where the ions are produced, for exact resonance $(\Delta_0 = 0)$ and for a detuning $\Delta_0 \approx 70 \text{ cm}^{-1}$. At a large detuning from resonance $(\Delta_0 \approx 70 \text{ cm}^{-1})$ the $N_i(Q)$ and, consequently, the W(F) dependences take the form of power laws with exponent $k = 4.2 \pm 0.2 = k_0$, in full agreement with our earlier measurements.^[9] At exact resonance between the energy of three quanta and the energy of the unperturbed 4f level, the W(F) dependence deviates appreciably from power-law. A rough approximation of this dependence by a power law, carried out by us for the purpose of illustration, yielded an exponent k = 1.5 \pm 0.3. It should be noted that saturation of the ion signal is observed in both cases. Figure 9 shows the results for different detunings. To make a comparison possible, the W(F) dependences were approximated by a power law. As seen from these results, there is a considerable detuning interval in which the dependence deviates from a power law with $k = k_0 = 4$. Thus, the results of the experiment show that when resonance sets in between the energy of several quanta and the energy of an isolated atomic level, the character of the multiphoton ionization is significantly altered. In particular, the onset of resonance is one of the reasons why the dependence of ionization probability on the radiation intensity deviates considerably from W ~ F^{k_0} .

An additional analysis of the experimental results, and particularly of the W(F) dependence at exact resonance, of the detuning interval in which the W(F) dependence differs from $W \sim F^{k_0}$, and of the radiation intensities at which identical ion signals were observed at $\Delta_0 = 0$ and $\Delta_0 = 70 \text{ cm}^{-1}$, enables us to draw a number of conclusions concerning the character of the resonantionization process. We turn first to the form of the W(F) dependence at exact resonance. Calculations show^[13] that under the conditions of our experiment the transition of an electron from the state 4f into the continuous spectrum is saturated. Consequently, the ionization process at exact resonance is determined by the process of three-photon excitation of the atom to the 4f



FIG. 8. Dependence of ion signal N_i on the energy in the radiation pulse Q: a-exact static resonance between the energy of three radiation quanta and the energy of the 4f level; b-deviation from resonance $\Delta_0 \approx 70 \text{ cm}^{-1}$.

state. The W(F) observed at exact resonance, which differs appreciably from $W \sim F^3$, can be due to the Stark shift of the resonant level with increasing radiation field. The reduced radiation line width for threephoton excitation is $\Gamma_{sh\omega} \sim 5 \text{ cm}^{-1}$. In measurement of the W(F) dependence, the field intensity ranged from 5×10^5 to 2×10^6 V/cm, and accordingly the energy of the 4f level changed by an amount from 1 to 10 cm^{-1} relative to the static value (see Sec. 3). Since ΔE_{4S-4f} $> \Gamma_{\rm sh\omega}$, detuning from resonance sets in with increasing radiation intensity, and this slows down the growth rate of the three-photon excitation probability in com-parison with the $W \sim F^3$ law. As seen from the calcu-lation results,^[6] to determine the concrete form of the W(F) dependence it is necessary to have exact data on the shape of the reduced emission line and on the value of the Stark shift. In addition, it is necessary to take into account the inhomogeneity of the radiation distribution in the region where the ions are produced, and the Stark level of the 4f level depends on the point in the radiation-focusing region at which the given atom is located. (During the time of action of the radiation pulse, $\tau \sim 10^{-8}$ sec, the displacement of atoms having practically thermal velocities is smaller than the diameter of the effective focusing circle, so that the atoms can be regarded in first approximation as immobile.) The resonant level of the atoms located in the field of the laser radiation becomes inhomogeneously broadened.^[13] A calculation based on the experimental data on the spectral and spatial distributions of the radiation



FIG. 9. Exponent in the power law $W = \alpha F^k$ vs. the deviation Δ_0 from resonance between the energy of three quanta and the energy of the unperturbed level 4f in the potassium atom.

has shown that the smoothing effect due to the inhomogeneous broadening of the 4f level is offset by the strong power-law dependence of the ionization probability on the radiation intensity (W $\sim F^3$). The results of this calculation are shown in Fig. 10. The solid curves show the atom distribution connected with the inhomogeneous broadening of the field in the ionization space, with respect to the Stark shift of the 4f level, for three different values of the field intensity (the maximum field of each measurement is given). These field values were realized in the measurement of the W(F) dependence and pertain to the beginning, midpoint, and maximum point of the range of variation of the intensity F. The ordinates (Fig. 10) represent the value of Neff, which is equal to the relative contribution of the different field regions to the total number of ionized atoms (Nx.eff ~ $N_{x,o}(F_x/F_{max})^k$). As seen from Fig. 10, the higher the field intensity the larger the broadening. Figure 10d shows the reduced radiation line shape for the threequantum process. Comparing Figs. 10a, b, c, and d, we see that the contribution of the regions in which the atoms with strongly shifted 4f line are located to the total number of ions decreases with increasing field, since the effective intensity is decreased as a result of the emission line shape. The altered effectiveness of the contributions of the different regions as a result of emission line shape is shown dashed in Figs. 10a. b. and c. It is clearly seen from Fig. 10 that when the field intensity decreases the fraction of the atoms excited to the 4f state (the area under the dashed curve) decreases relative to the total number of atoms (area under the solid curve).

The foregoing analysis shows that in the case of an inhomogeneous field distribution in the ionization volume, the Stark shift of the resonant level can greatly alter the functional dependence of the ionization probability on the radiation intensity. A change in the functional dependence W(F) was observed not only in the



FIG. 10. Inhomogeneous broadening of resonant level as a result of the Stark shift in an electric field that is unevenly distributed in the radiation-focusing region. N_{eff} is the relative contribution of the different regions to the total number of produced ions. The solid and dashed curves are drawn without and with allowance for the emission line width. a, b, c-distributions at different field intensities; d-reduced emission line. case of exact resonance, but also near it, at detunings of -20 cm^{-1} (towards shorter wavelengths) and $+10 \text{ cm}^{-1}$ (towards longer wavelengths) from the position $\Delta_0 = 0$. We see that the detunings are asymmetrical relative to the static position of the 4f level. It can be assumed that such an asymmetry is connected with the sign of the Stark shift of the 4f level towards smaller wavelengths, as predicted by the theory.^[6,7] The absolute magnitude of the detuning, $\Delta \approx -20 \text{ cm}^{-1}$, corresponds to the change in the energy of the 4f level at the maximum radiation field intensity ($\Delta E_{4S-4f} \approx 20 \text{ cm}^{-1}$).

The absolute value of the cross section of threephoton resonant excitation of the potassium atom to the 4f state can be estimated by comparing the amplitudes of the ion signals at exact resonance and at large detuning (Fig. 8). Since we did not measure either the density of the atomic beam or the absolute sensitivity of the ion detector with sufficient accuracy, the absolute value of the cross section can be determined with sufficient accuracy by starting from the saturation of the ion signal.^[14,15] The cross section of four-photon ionization of the potassium atom was determined in this manner in ^[15] under conditions when no resonances are produced. The obtained value $\alpha_4 = 10^{-109}$ (the exponent with allowance for the errors is $109^{+1.6}_{-1.5}$), agrees well with the perturbation-theory calculation.^[15] Using the same value of α_4 and the ratio of the ion-signal amplitudes at $\Delta_0 \approx 70$ cm⁻¹ and $\Delta_0 = 0$ (Fig. 8), we can obtain the cross section for three-photon excitation. We take into account here the fact that the process of singlephoton ionization from the excited level is saturated and therefore the total number of ions at exact resonance is determined only by the probability of the threephoton excitation. As is well known, the number of ions is determined from the relation

$N_i = n_0 V_k \alpha_k F^k \tau_k.$

By measuring the intensities at which the number of ions is the same for the cases $\Delta_0 = 0$ and $\Delta_0 = 70$ cm⁻¹, we find that

$$\alpha_{3} = \frac{V_{4}}{V_{3}} \frac{\tau_{4}}{\tau_{3}} \frac{F_{\Delta_{0}=70}^{4}}{F_{\Delta_{0}=0}^{3}} \alpha_{4}.$$
(7)

In the procedure of calculating α_3 with the aid of this formula, we took into account the fact that, out of the total emission line width, the only photons that actually participate in the three-photon excitation process are those whose energy corresponds to the 4f level with allowance for its ionization width ($\gamma \sim 1 \text{ cm}$).^(6,7) The quantity α_3 determined in accordance with (7) is equal to^[16]

$$\alpha_{3 \exp} = 10^{-73} \operatorname{cm}^{6} \cdot \operatorname{sec}^{2}$$

(The exponent with allowance for the errors is 73^{+3}_{-3}). Perturbation-theory calculation^[6] yields for the conditions of our experiment

$$\alpha_{3 \text{ theor}} = 10^{-71} \text{ cm}^6 \cdot \text{sec}^2$$

(with an exponent 71^{+2}_{-3}). The agreement between the ex-

perimental and theoretical values of α_s can be regarded as satisfactory.

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A comparison of the cross section for the four-photon ionization of the potassium atom ($\alpha_4 = 10^{-109}$) with the cross section for three-photon excitation of the level ($\alpha_3 = 10^{-73}$) shows that in the case when the transition from the 4f state to the continuous spectrum is saturated, the resonant-ionization process requires a lower radiation intensity. It should be noted that the advantage of the resonant ionization process becomes larger the narrower the radiation spectrum and hence the larger the fraction of the photons that can lead to resonant excitation of the atom.

We have investigated the resonant multiphoton ionization of atoms jointly with B. A. Zon, N. L. Manakov, and Professor L. P. Rapoport of the Department of Theoretical Physics of the Voronezh University, whose studies^[6,7] exerted a strong influence on the interpretation of the experimental results.

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- ¹L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964) [Sov.
- Phys.-JETP 20, 1307 (1965)].
- ²N. D. Voropaev and A. N. Oraevskii, FIAN Preprint No. 33, 1966.
- ³G. S. Voronov, Zh. Eksp. Teor. Fiz. 51, 1496 (1966) [Sov.
- Phys.-JETP 24, 1009 (1967)].
- ⁴V. A. Kovarskii, Proc. 9 Int. Conf. Phen. Ioniz. Gases, vol. 32, Bucharest, 1969.
- ⁵N. V. Lebedev, Opt. Spektrosk. **28**, 1026 (1970); Opt. Spektrosk. **30**, 381 (1971).
- ⁶B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. **60**, 1264 (1971) [Sov. Phys.-JETP **33**, 683 (1971)].
- ⁷V. A. Davydkin, B. A. Zon, N. L. Manakov, and L. P. Rapoport, Zh. Eksp. Teor. Fiz. **60**, 124 (1971) [Sov. Phys.-JETP **33**, 70 (1971)].
- ⁸N. B. Delone and L. V. Keldysh, FIAN Preprint No. 11, 1970.
- ⁹G. A. Delone, N. B. Delone, N. P. Donskaya, and K. B. Petrosyan, Zh. Eksp. Teor. Fiz. Pis'ma Red. 9, 192 (1969) [JETP Lett. 9, 112 (1969)]; Proc. 9 Int. Conf. Phen. Ioniz. Gases, vol. 30, Bucharest, 1969.
- ¹⁰P. Agostini, G. Barjot, G. Mainfray, and I. Morellec, Dig. Techn. Pap. 6 Int. Quat. Electr. Conf., 92, Kioto, Japan, 1970.
- ¹¹G. A. Delone and N. B. Delone, Zh. Eksp. Teor. Fiz. Pis'ma Red. 10, 413 (1969) [JETP Lett. 10, 265 (1969)].
- ¹²T. M. Barkhudarova, G. S. Voronov, V. M. Gorbunkov, G. A. Delone, N. B. Delone, and N. K. Martakova, Paper at Conf. International Radio Union on Laser Measurements, Warsaw, 1968; G. A. Delone, and M. S. Rabinovich, School Lectures on the Physics of Ionized Gases, Herceg-Novi, Yugoslavia, 1968.
- ¹³G. A. Delone, Dissertation, FIAN, 1971.
- ¹⁴N. K. Berezhetskaya, G. S. Voronov, G. A. Delone, N. B. Delone, and G. K. Piskova, Zh. Eksp. Teor. Fiz. **58**, 753 (1970) [Sov. Phys.-JETP **31**, 403 (1970)].
- ¹⁵G. A. Delone, N. B. Delone, and G. K. Piskova, FIAN Preprint No. 62 (1971).
- ¹⁶G. A. Delone, N. B. Delone, and G. K. Piskova, FIAN Preprint No. 60 (1971).

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