MULTIPHOTON IONIZATION OF ATOMS. II. IONIZATION OF KRYPTON BY RUBY-LASER RADIATION

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Multiphoton ionization of the krypton atom (J = 13.996 eV) by radiation from a ruby laser ($\hbar\omega$ = 1.785 eV) was investigated experimentally. It was observed at a photon flux $F \sim 10^{31}$ cm^{-2} sec^{-1}, corresponding to an electric field strength $E \sim 3 \times 10^7$ V/cm. Measurements of the ionization probability and its dependence on the photon flux are described. The ionization probability is proportional to the photon flux to the power $K \approx \langle J/\hbar\omega + 1\rangle - 2$. An analysis of the experimental and theoretical results indicates that transitions between bound states contribute appreciably to the ionization probability and that the effect of the radiation field on these states is considerable.

INTRODUCTION

MULTIPHOTON ionization by ruby-laser emission ($\hbar\omega$ = 1.785 eV) was observed recently for xenon (J = 12.127 eV)[1], krypton (J = 13.996 eV), and argon (J = 15.755 eV)[2] atoms at a photon flux intensity $F \approx 10^{30}$ cm^{-2} sec^{-1}, corresponding to an electric field intensity $E \approx 10^7$ V/cm. For xenon[1], measurements were made of the ionization probability and of the dependence of the probability on the photon flux intensity. For krypton and argon[2], measurements were made of the ratio of the ion signals to the signal from the xenon, making it possible to estimate the relative values of the ionization probability.

The ionization of an atom in a strong electromagnetic field with quantum energy $\hbar\omega < J$ was considered theoretically by L. V Keldysh[3], who obtained a formula for the probability of ionization as a result of transitions between virtual levels of the continuous spectrum, with exact account taken of the effect of the field on these levels. The numerical calculations of the probability of multiphoton ionization of atoms of hydrogen and noble gases, obtained in perturbation theory up to 14th order, are presented in the papers of Gold and Bebb[4,5].

The present paper is devoted to an investigation of multiphoton ionization of krypton by ruby-laser emission. We measured the absolute ionization probability and its dependence on the intensity of the photon flux at $F \approx 10^{31}$ cm^{-2} sec^{-1}.

2. EXPERIMENT

The experimental setup was in the main similar to that used by us earlier in the investigation of multiphoton ionization of xenon[1]. The emission from a powerful ruby laser was focused into a vacuum chamber filled with krypton at 10^{-4} mm Hg, when the mean free path (~ 40 cm) and the time between collisions (~ 10^{-3} sec) are larger by several orders of magnitude than the dimensions of the focusing region (~ 10^{-2} cm) and the duration of the radiation pulse (~ 10^{-8} sec). Thus, ionization is the result of the direct action of the radiation on the individual atoms.

The mean free path of electrons of energy ~ 1 eV in the plasma that is produced in the focal region and has a density ~ 10^{12} cm^{-3} (assuming 100% ionization) is of the order of 1 cm, which is much larger than the dimension of the focusing region (~ 10^{-2} cm), so that there are likewise no electron-ion collisions in the ionization region.

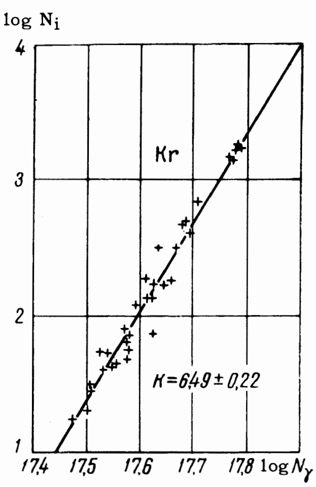
A. Measurement of the dependence of the ionization probability on the photon flux intensity. To determine the dependence of the ionization probability on the photon flux intensity we made relative measurements of the dependence of the number of produced ions on the number of photons passing through the focusing region at a constant space-time distribution of the focused laser emission[1].

Constancy of the space-time distribution of the emission was ensured by thorough stabilization of the operating conditions of the laser and was monitored against the multiphoton ion signal. The emission energy in the giant pulse was constant during the time of the experiment within ~2%. The emission intensity can change even if the emission energy is constant, owing to the changes in the space-time distribution of the emission. The scatter in the values of the control multiphoton signal from xenon has shown that the instability of the photon flux intensity was ~5%.

The radiation was attenuated with two thick glass plates operating in reflection. The plates were placed at angles α and $180°-\alpha$ to the radiation-beam axis, so that the shift of the beam due to refraction in the first plate was compensated for by the second plate. The plates were thick enough so that no interference was produced. The attenuation coefficient was varied by changing the angle α. The plate rotation axis was in the polarization plane of the radiation.

A typical plot of the number of produced krypton ions N_i against the number of photons N_γ passing through the focusing region is shown in a log-log scale in the figure. To determine the dependence of the ionization probability on the photon flux intensity we drew, in accord with the expected relation $W = AF^K$, a straight line $\log N_i = K \log N_\gamma + C$ through the experimental points by the method of least squares. The results of a series of experiments are listed in Table I.

The weighted mean for all the experiments was



Dependence of the number of produced krypton ions on the number of photons emitted from a ruby laser and passing through the focusing region (in relative units).

Table I. Results of a series of experiments on the measurement of K.

Experiment No.	K	Experiment No.	K
1	5.86±0.44	5	6.16±0.33
2	6.41±0.37	6	6.14±0.71
3	6.48±0.21	7	6.78±0.39
4	6.49±0.29	8	5.92±0.23

$K = 6.31 \pm 0.11$. The quoted error is equal to the dispersion of the weighted arithmetic mean value of K, calculated from the scatter of the values of K in individual experiments. The dispersion of the distribution of the values of K was 0.28. The average dispersion, obtained from the dispersions of the individual measurements listed in Table I, is 0.37. A comparison of these quantities show that the scatter and the results of individual measurements of K is strictly statistical.

The absolute measurement of the photon flux intensity gives for the intensity in the center of the interval of the measurements a value $F = 4.8 \times 10^{30}$ cm^{-1} sec^{-1}, corresponding to $E = 2.2 \times 10^7$ V/cm. The measurement interval was $(3.2-6.4) \times 10^{30}$ cm^{-2} sec^{-1}.

B. Measurement of the absolute value of the ionization probability. To determine the absolute value of the ionization probability $W = N_i/nV_K\tau_K$ to the absolute value of the photon flux intensity $F = N_\gamma/S\tau$, it is necessary to carry out absolute measurements of the density n of the neutral atoms, the number N_i of the produced ions, and the number N_γ of the photons passing through the focusing region, and also to measure the space-time distribution of the focused radiation, from which the values of S, τ, V_K, and τ_K are calculated (see[1]). The physical meaning of the effective volume V_K and of the effective duration τ_K for the multiphoton ionization process lies in the fact that the contribution of the elementary volume dV to the total number of ions produced in the time dt is proportional to the relative intensity at the given point and at the given instant of time, raised to the power K. The steeper the dependence of the ionization probability on the intensity of the photon flux, the smaller the effective volume and the effective time.

The time distribution of the radiation was investigated with a photocell having a total resolution < 1 nsec. Within the solid angle ~10^{-5} sr bounded by the entrance aperture of the objective, the distribution had the form of a smooth bell-shaped curve with $\tau = 35.4 \pm 0.4$ nsec. We measured also the time distribution in the regions where the intensity of the focused radiation was maximal. To this end we focused on the photocell the radiation bounded by a diaphragm located in the focus of a long-focus lens (f = 2 m). The dimension of the diaphragm was chosen smaller than the dimension of the individual maximum-intensity regions[6]. In this case the distribution also had the form of a smooth bell-shaped curve with $\tau = 37.0 \pm 065$ nsec, which did not differ in practice from the duration of the distribution of the entire flux. This confirms the possibility of separating the spatial and the temporal variables when measuring the space-time distribution of the radiation (see[1]). The magnitude of the effective time for the experimentally measured quantity K = 6.3 was τ_K = 13.7 nsec.

The spatial distribution of the focused radiation was measured by a photometric method[1,6]. The minimum cross section area, normalized to the maximum photon flux intensity, was $S = (2.85 \pm 0.8) \times 10^{-6}$ cm^{-2}. The value of the effective value for the experimentally measured quantity K = 6.3 was $V_K = 10^{-7.7 \pm 1.2}$ cm^3.

The experiment was carried out at a neutral atom density $n = 1.2 \times 10^4$ cm^{-3}. The density was determined by measuring the pressure with an ionization manometer, with due allowance for the dependence of its sensitivity on the type of gas[7]. The accuracy of the absolute graduation of the manometer was ~ 50%.

The absolutization of the sensitivity of the electron multiplier was carried out earlier for xenon[1] with the aid of a Faraday cylinder. The same data were used for the krypton, taking into consideration the dependence of the secondary-emission coefficient of the first dynode on the mass of the registered ion[2]. Allowing for the statistical error, the number of the registered ions was $N_i = 10^{3.8 \pm 0.5}$.

The absolute number of photons N_γ passing through the focusing region was determined by measuring the emission energy with a commercial calorimeter (type IMO-1) placed behind the chamber. The accuracy of the absolute calibration of the calorimeter was 15%. We took into account the correction for the reflection of the radiation from all the surfaces lying in the path of the light from the focusing region to the calorimeter. The value of N_γ was $10^{17.8 \pm 0.18}$.

The absolute value of the probability of the multiphoton ionization of krypton, calculated from this data by means of the formula given above, was $W = 10^{6.3 \pm 2.4}$ sec^{-1} at a photon flux intensity $F = 10^{30.8 \pm 0.3}$ cm^{-2} sec^{-1}.

3. DISCUSSION OF RESULTS

The experiment has shown that for $F \approx 10^{30}$ cm^{-2} sec^{-1} ($E \sim 10^7$ V/cm) the power in dependence of the probability of the multiphoton ionization of krypton on the intensity of the photon flux is much lower than the number of quanta necessary for the ionization, $K_0 = \langle J/\hbar\omega + 1\rangle$, where $\langle x\rangle$ denotes the integer part of x. A qualitatively similar result was obtained earlier for xenon[1]. These data allow us to assume that application of a field of intensity $E \sim 10^7$ V/cm greatly distorts the states of the atom.

The strongest effect is exerted by the radiation field on the states of the continuous spectrum. However, as shown by Keldysh's theory[3], in which this action is taken exactly into account, in those regions of field intensity and frequency values where the ionization process has a multiphoton character, we have $W \sim F^{\langle J/\hbar\omega + 1\rangle}$, i.e., $K = K_0$. The power in the W(F) dependence changes on going into the region of the tunnel effect. The slope of the W(F) curve increases, and the dependence becomes exponential.

The decrease in the slope of W(F) can thus be due only to the effect of the radiation field on the bound states of the atom.

The action of the radiation field causes a shift in a broadening of the levels, as a result of which the upper levels overlap and merge practically into a continuous spectrum adjacent to the spectrum of the free states. This situation is analogous to the lowering of the effective ionization potential of the atom. According to estimates by L. V. Keldysh[8], for a hydrogenlike atom the ionization potential is lowered by an amount $\Delta J \sim (ea_0E)^{2/5}J^{3/5}$, which gives $\Delta J \sim 1$ eV for $E \sim 10^7$ V/cm. The value $K \approx (K_0 - 1)$ observed experimentally for xenon can be attributed to this mechanism[1]. In the case of krypton, the observed value is $K \approx (K_0 - 2)$, which corresponds to a lowering of the effective ionization potential by 3.5 eV. At such a depth, the distances between the energy levels are already several tenths of an electron volt. According to the estimate given by L. V. Keldysh, the field intensity required to lower the effective ionization potential by 3.5 eV is $E \approx 10^8$ V/cm. Therefore the value of K observed for krypton is apparently connected not with the lowering of the effective ionization potential alone.

If the spectrum of the atom contains quasi-resonant levels whose energies relative to the ground state are close to the energy of an integral number of radiation quanta, then the contribution of the transitions between these levels to the ionization probability can be larger than the contribution of the transitions between the virtual levels of the continuous spectrum[3]. The broadening and the energy shift of the quasi-resonant level by the radiation field greatly influence the ionization probability and its dependence on the photon flux intensity[9]. A calculation of this effect, in an approximation in which the broadening and the shift of the level in the radiation field are comparable with the resonance detuning for only one quasi-resonant level, gives for xenon and krypton a value of K which coincides with experiment at the higher-

Table II. Probability of multiphoton ionization.

	Experiment	Formula of L. V. Keldysh[8]	Calculation of Bebb and Gold [5]
W, sec^{-1}	$10^{6.3\pm2.4}$	$10^{0.3\pm2.4}$	$10^{13.7\pm2.4}$

intensity borderline of the experimental error. Such a deviation is apparently connected with the fact that the calculation did not take into account other levels and the overlap of the upper levels in the strong field.

The experimental value of the probability of the multiphoton ionization and the results of calculations by the formula of L. V. Keldysh (see[8] as well as formula (4) in[1]) and by perturbation theory[5] are given in Table II for $F = 10^{30.81\pm 0.3}$ cm^{-2} sec^{-1}. The errors in the calculated values were obtained with allowance for the experimental accuracy with which F is measured.

The experimentally observed ionization probability exceeded the calculated probability of the transitions between virtual levels of the continuous spectrum, this being apparently due to the contribution of the transitions between bound states. The perturbation-theory calculation gives too high a value for the ionization probability, for no account is taken here of the broadening of the levels.

The aggregate of the experimental data on multiphoton ionization of xenon[1] and krypton shows that the transitions between bound states make an appreciable contribution to the ionization probability and that the action of the radiation field on these states is appreciable.

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[1] G. S. Voronov and N. B. Delone, JETP **50**, 78 (1966), Soviet Phys. JETP **23**, 54 (1966).

[2] G. S. Voronov, G. A. Delone, and N. B. Delone, JETP Letters **3**, 480 (1966), transl. p. 313.

[3] L. V. Keldysh, JETP **47**, 1945 (1964), Soviet Phys. JETP **20**, 1307 (1965).

[4] A. Gold and B Bebb, Phys. Rev. Lett. **14**, 60 (1965).

[5] B. Bebb and A. Gold, Phys. Rev. **143**, 1 (1966).

[6] T. M Barkhudarova, G. S. Voronov, V. M. Gornunkov, and N. B. Delone, JETP **49**, 386 (1965), Soviet Phys. JETP **22**, 269 (1966).

[7] R. Barre, R. Geller, and G. Mongonin, Le Vide **69**, 195 (1957).

[8] G. S. Voronov, V. M. Gorbunkov, G. A. Delone, N. B Delone, L. V. Keldysh, M. S. Rabinovich, Paper at 7th Intern. Conf. on Ionization Phenomena in Gases, Belgrade, 1965; FIAN Preprint A-115, 1965.

[9] G. S. Voronov, JETP **51**, 1496 (1966), Soviet Phys. JETP **24**, 1009 (1967).

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