MANY-PHOTON IONIZATION OF THE XENON ATOM BY RUBY LASER RADIATION

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Submitted to JETP editor August 27, 1965

J. Exptl. Theoret. Phys. (U.S.S.R.) 50, 78-84 (January, 1966)

A description is given of experiments on the many-photon ionization of a xenon atom ($I = 12.13$ eV) in the strong electric field of the radiation from a ruby laser ($\hbar\omega = 1.79$ eV). In a field of $E \sim 10^7$ V/cm intensity, the effectiveness of the ionization during a period of $\approx 10^{-8}$ sec was proportional to the photon beam intensity raised to the power ~ 6. Reasons are given why this quantity may be less than $\langle I/\hbar\omega + 1\rangle$. The many-photon ionization theory[1] gives, for the six-photon process, a dependence of the ionization probability on the electric field intensity which is in order-of-magnitude agreement with the experimental results. The same value of the ionization probability is given by a numerical calculation using the perturbation theory and allowing for the resonance levels in the xenon spectrum.[7]

1. INTRODUCTION

KELDYSH[1] showed that the many-photon ionization and the tunnel effect are the limiting cases of the same process of the ionization of an atom in an alternating electric field. In a strong field, whose frequency is such that an electron is still able to cross a potential barrier during the period of the field, the ionization process is similar to the usual tunnel effect and the dependence of the ionization probability per unit time on the field intensity is exponential. When the frequency is higher or the field intensity lower than in the previous case, this dependence is of the power type $W = AF^k$ (where F is the photon beam intensity), which is characteristic of the many-photon processes. Since the barrier width is $\sim I/eE$, and the electron velocity is $\sim\sqrt{I/m}$, the critical frequency is $\omega_{cr} = eE/\sqrt{2mI}$. When $E \sim 10^7$ V/cm and $I \sim 10$ eV, $\omega_{cr} = 10^{14}$ sec^{-1}.

It has recently become possible to produce strong alternating electric fields by focusing the radiation of a powerful laser. The energy of the radiation quanta of such a laser is about an order of magnitude less than the ionization potential of the majority of atoms and molecules and the frequency $\sim 10^{15}$ sec^{-1} exceeds ω_{cr} up to fields $E \sim 10^8$ V/cm. Consequently, in fields $E < 10^8$ V/cm, we may expect the ionization to be of the many-photon type.

An estimate made on the basis of Keldysh's work[1] shows that, when $\omega \sim 10^{15}$ sec^{-1}, an electric field of $\sim 10^7$ V/cm intensity, which corresponds to a photon beam intensity $\sim 10^{30}$ cm^{-2} sec^{-1}, is needed to reach the experimentally observed probability of the many-photon ionization. To reach this intensity, one may use the giant pulses of ruby and neodymium-glass lasers and, possibly, their harmonics.

The many-photon ionization of the xenon atom[2] and of the hydrogen molecule[3] by the radiation from a ruby laser has already been observed experimentally. The present paper describes an investigation of the dependence of the effectiveness of the ionization on the photon beam intensity and the measurement of the absolute value of the average probability of ionization per unit time.

2. EXPERIMENTAL ARRANGEMENT

In these experiments we used xenon, which has the lowest ionization potential I, equal to 12.13 eV, among the monatomic gases. We used a ruby laser Q-switched by a Kerr cell. The radiation was focused by an objective with a focal length $f' = 27$ mm, corrected for spherical aberration at $\lambda = 6940$ Å (Fig. 1). The measurements were carried out at a pressure lower than 10^{-3} mm Hg, when the mean free path (~ 1 cm) was several orders of magnitude greater than the region in which the radiation was focused ($\sim 10^{-2}$ cm) and the time between collisions ($\sim 10^{-6}$ sec) was longer than the duration of the laser radiation pulse ($\sim 10^{-8}$ sec).

The average probability of ionization per unit time W was determined by measuring the number of ions N_i formed by a single laser radiation pulse, the effective volume V_k in which radiation of sufficient intensity was concentrated, and the

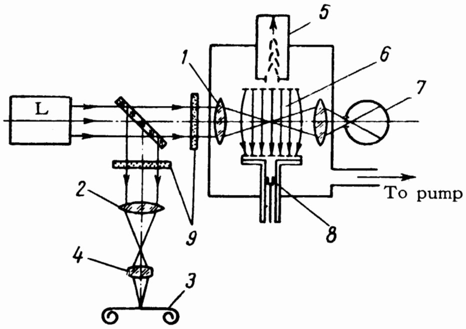

FIG. 1. Experimental arrangement. The many-photon ionization took place in a vacuum chamber under the action of radiation from a laser L, focused by an objective 1. Some of the laser radiation was split off to pass through an objective 2, identical with, and at the same distance from the laser as, the objective 1.[4] The spatial distribution of the radiation focused by the objective 2, identical with the distribution of the radiation focused in the camera, was photographed on an enlarged scale on a film 3 through a microscope objective 4. 5 is an electronic multiplier, 6 – an electric field for extracting the ions, 7 – a calorimeter, 8 – a Faraday cup, and 9 – attenuators.

effective duration of action of the field τ_k. The number of recorded ions is

$$N_i = n \int W\,dV\,dt = An \int F^k\,dV\,dt,$$

where n is the density of neutral atoms. The intensity of the photon beam varied widely from point to point in the region where the laser radiation was focused.[4] The complexity of the spatial structure of the focused laser radiation and the strong dependence of the probability of the many-photon ionization on the photon beam intensity led to the ionization taking place in numerous regions near points with the highest field intensity. The value of the effective volume of each such region depended on the rate of decrease of the ionization probability with the decrease in the field intensity, i.e., depended on the value of k, which is the number of quanta whose absorption probability governed the ionization probability.

When a laser is operating under Q-switching conditions, the radiation conditions are satisfied simultaneously, at the moment of Q-switching, for many oscillation modes. The threshold values of the Q-factor for the various modes differ by an amount which is considerably less than that by which the Q-factor is altered by the modulation. Therefore, the generation of the various modes proceeds at approximately the same rate and the radiations of these modes are superimposed in time. In our apparatus, the time distribution of the radiation, measured with a photocell of $\sim 10^{-9}$ sec resolution, was a smooth curve with a half-width of $\sim 3 \times 10^{-8}$ sec.[1)]

Therefore, the function describing the space-time characteristics of the radiation may be represented in the form

$$F(x, y, z, t) = F_\gamma \psi(x, y, z)\varphi(t);$$

F_γ is found from the normalization:

$$N_\gamma = F_\gamma \int\int \psi(xy)\varphi(t)\,dS\,dt = F_\gamma S\tau,$$

where N_γ is the total number of photons which have passed through the focusing region, $S = \int \psi(xy)dS$ is the effective cross-section area of the focused radiation, and $\tau = \int \varphi(t)dt$ is the effective duration of the radiation.

The number of ions formed, N_i, is related to the number of photons, N_γ, by

$$N_i = An \frac{V_k \tau_k}{(S\tau)^k} N_\gamma^k, \tag{1}$$

where V_k and τ_k are the values of the effective volume and time for a k-quantum process,

$$V_k = \int \psi^k(x, y, z)\,dV,$$

$$\tau_k = \int \varphi^k(t)\,dt.$$

From the measured dependence of N_i on N_γ, we can determine the value of k, if all the other quantities in Eq. (1) remain constant. To determine the absolute values of the ionization probability

$$W = \frac{N_i}{nV_k\tau_k} \tag{2}$$

and of the photon beam intensity

$$F = \frac{N_\gamma}{S\tau} \tag{3}$$

the values of V_k, τ_k, S, τ were calculated from the measured distribution functions $\psi(x, y, z)$ and $\varphi(t)$.

A Faraday cup and an electronic multiplier were used as ion detectors. The sensitivity limits were 10^3 and 10 ions, respectively. To eliminate the background of ions generated by the intense beam of light on the surfaces of various solids, such as the objective lenses, we used a weak ($\sim$10 V/cm) electric field which attracted the ions to a detector, in front of which was a system of diaphragms. In this way, we were able to reduce the ion background by many orders of magnitude so that the signal due to these ions was of the same level as the sensitivity limit of the apparatus. (We

[1)]See the note added in proof.

may assume that in [5], where an open probe was used, this background was recorded.) The mass control was carried out during the time of flight of ions to a detector, which was placed at distances of from several centimeters to a meter from the region of focused radiation.

The total number of photons N_γ, that had traversed the focusing region was measured with a calorimeter. The total number of photons was varied by means of attenuating filters, whose attenuation linearity was checked experimentally. This method of varying N_γ did not alter the space-time distribution of the radiation.

The distribution of the focused radiation in space $\psi(x, y, z)$ was measured by a photoelectric method using photographs of various cross sections of the focused radiation, obtained on an enlarged scale using a microscope objective. As a result of the measurements carried out on a number of cross sections located at various distances from the focal plane of the objective, we collected data on the volume which contained radiation of given intensity.

The function giving the distribution of the radiation in time $\varphi(t)$ was measured with a photocell whose resolution was better than 10^{-9} sec.

3. RESULTS OF EXPERIMENTS

Figure 2 gives the experimental data on the dependence of the number of ions, formed as a result of the many-photon ionization of xenon, on the number of photons which have passed through the focusing region during one laser pulse. The experimental data are represented approximately, in accordance with Eq. (1), by a straight line $\log N_i = \log C + k \log N_\gamma$ found by the least-squares method. It was thus found that $k = 6.23 \pm 0.14$.

The scatter of the values of N_i for $N_\gamma = \text{const}$ was due to the different values of the distribution function $\psi(x, y, z)$ for the different laser radiation pulses. The form of the function $\psi(x, y, z)$ in each actual radiation pulse depended on a number of factors which govern the operation of the laser, only some of which can at present be controlled and stabilized. Measurements of the function $\psi(x, y, z)$ for many laser pulses, using the photometric method, showed that although the actual form of $\psi(x, y, z)$ was not constant, the average characteristics of the radiation distribution (for example, $S = \int \psi(x, y)\,dS$) did not change greatly. The photometric data gave $S = (6 \pm 1.6) \times 10^{-6}\ \text{cm}^2$. The error in the value of S was in agreement with the scatter of the value of N_γ for $N_i = \text{const}$ (cf. Fig. 2). Measurements of the $\varphi(t)$ function show

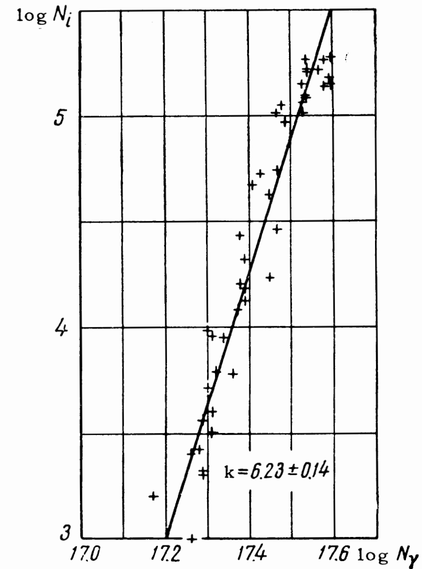

FIG. 2. Dependence of the number of ions formed N_i on the number of photons N_γ which have traversed the focusing region.

that this function was practically constant, the effective duration of the radiation pulse being $\tau = \int \varphi(t)\,dt = 3.1 \times 10^{-8}$ sec.

Control experiments showed that the true error in the value of k exceeded the reported statistical error. The true error was associated with a systematic variation of the focused laser radiation distribution in space, which was difficult to allow for. At present, it is difficult to indicate the value of the true error; we may assume that it is several times greater than the statistical error.

The absolute value of the photon beam intensity in the middle of the range of measurements was $F = 10^{30.25 \pm 0.25}\ \text{cm}^{-2}\ \text{sec}^{-1}$, which was equivalent to an electric field intensity $E = (1.3 \pm 0.3) \times 10^7$ V/cm.

Using the most probable value of k obtained experimentally, we calculated the value of the effective volume $V_6 = \int \psi^6(x, y, z)\,dV$ in order to find the absolute value of the ionization probability. From the photometric measurements, we found that $V_6 = 2.5 \times 10^7\ \text{cm}^3$. The value of V_6 was very sensitive to a change in $\psi(x, y, z)$ and therefore the error in obtaining its absolute value reached several hundred per cent. The effective time was $\tau_6 = 1.1 \times 10^{-8}$ sec.

The errors in finding the absolute value of all the quantities in Eq. (2) were considerably less than those in V_6. A calculation using Eq. (2) gave the most probable value of the ionization probability $W = 10^{5.5 \pm 1.7}\ \text{sec}^{-1}$ for the photon beam intensity $F = 10^{30.25 \pm 0.25}\ \text{cm}^{-2}\ \text{sec}^{-1}$ ($E = (1.3 \pm 0.3) \times 10^7$ V/cm). The experimental data, in the form

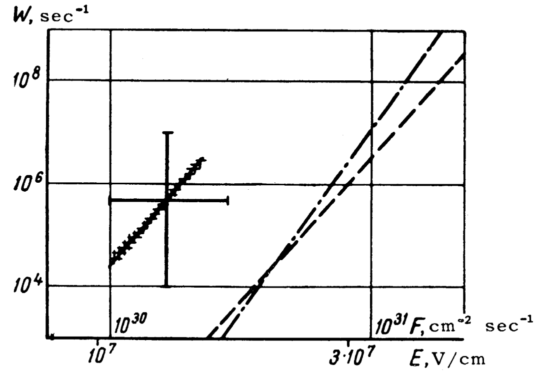

FIG. 3. Dependence of the probability of the many-photon ionization of the xenon atom on the intensity of the photon beam (the electric field intensity) for $\hbar\omega = 1.79$ eV. The results of calculation are also given: the dashed line represents the calculations made in accordance with [1] for the six-photon process; the chain line represents the calculations made in accordance with [7] allowing for resonance levels.

of the ionization probability on the photon beam intensity (electric field intensity), are given in Fig. 3.

The experiments were carried out using a neutral xenon-atom density $n = 2 \times 10^{13}$ cm^{-3}. Comparison of the values of nV_k and N_i showed that in our experiments the effectiveness of the ionization was several per cent.

4. DISCUSSION OF EXPERIMENTAL RESULTS

From a comparison of the ionization potential of xenon I, equal to 12.13 eV, and of the energy of the ruby laser radiation quanta $\hbar\omega = 1.79$ eV, it is evident that seven quanta are needed to produce the ionization. The most probable experimental value of k is 6.23. The discrepancy may be due to the effect of an electric field $\sim 10^7$ V/cm on the upper electron levels in the xenon atom.[6] The higher the levels are, the higher the density and the greater the Stark shifts. When the level shifts become of the same order as the distances between the levels, intense transitions from level to level begin to take place in the laser-beam field. Consequently, the levels broaden strongly and practically merge into a continuous spectrum, adjacent to the spectrum of free states.

An electron falling onto one of these levels will, after a fairly long time, have the near certainty of being lost to the atom. For the hydrogen atom, the separation between the higher levels is of the order of $(\epsilon_S^3/I)^{1/2}$, where S is the number of the level, and its shift is $eEa_0S^2 \sim eEa_0I/\epsilon_S$, where a_0 is the Bohr radius. A comparison of these two quantities shows that the boundary of the continuous spectrum will shift by $\epsilon \sim (eEa_0)^{2/5}I^{3/5}$, i.e., by ~ 1 eV in a field $E \sim 10^7$ V/cm. This order-of-magnitude estimate is valid for other atoms for sufficiently high, and therefore hydrogen-like, levels.

According to Keldysh's theory,[1] the probability of the many-photon ionization at $\omega \sim 10^{15}$ sec^{-1} in $E < 10^8$ V/cm can be calculated from the formula

$$W = \omega\left(\frac{I}{\hbar\omega}\right)^{2l+3/2}\left(\frac{e^2E^2}{3m\omega^2 I}\right)^{\langle I/\hbar\omega+1\rangle}, \tag{4}$$

where $\langle x \rangle$ represents the integral part of x, l is the orbital electron momentum, and I is the effective ionization potential. Calculation, using Eq. (4) for the six-photon process, gives the experimentally observed value of the probability $W = 10^{5.5}$ sec^{-1} for an electric field intensity $E = 2.7 \times 10^7$ V/cm. The same value of the field intensity was obtained in [7] where a numerical calculation was reported using the seventh-order perturbation theory with allowance for the resonance levels.

In the xenon atom, the resonance level may be the level 7p $^3S_1$, whose energy, relative to the ground state, differs by 0.188 eV from the energy of six quanta, or the level 6s $^3P_0$, whose energy differs by 0.219 eV from the energy of five quanta. In view of the present state of the theory and experimental knowledge, we may regard the results to be in sufficiently good agreement.

It does not yet seem possible to draw any final conclusions about the many-photon ionization mechanism. Experiments are needed using other atoms, frequencies and radiation intensities.

The authors are grateful to V. S. Karyukhin, S. A. Ob''edkov, and V. P. Solov'ev for their help in this investigation, and to G. A. Delone, L. V. Keldysh, and M. S. Rabinovich for their valuable comments.

Note added in proof on December 1, 1965. In some cases[8], various parts of a ruby crystal are not uniformly de-excited in a giant pulse. The duration of radiation of a given region is found to be 1.5-2 times shorter than the duration of the crystal rod end as a whole. Under such conditions, the true value of the electric field intensity may exceed the value calculated from the duration of radiation from the whole crystal by a factor not greater than 1.5.

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Translated by A. Tybulewicz
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