MEASUREMENT OF LIFETIMES OF EXCITED STATES OF THE HYDROGEN ATOM

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A method is described of determining lifetimes of atoms in excited states by observing the increase in the intensity of light radiated by a beam of fast atomic particles as it traverses a gaseous target. The lifetimes of hydrogen atoms in states with n = 3, 4, and 5, produced in the dissociation of H_2^+ ions (energy range 10—30 keV) in He, have been measured by using this method. The values obtained for these lifetimes are in good agreement with those calculated by quantum mechanics for a statistical population of the fine structure sublevels of the hydrogen atom.

1. INTRODUCTION

IN measurement of excitation cross sections of energetic ions or atoms in collisions with gas molecules, we must consider the following fact. In the apparatus usually used for this type of experiment, a monoenergetic beam of the ions or atoms being studied enters a collision chamber filled with gas to a pressure at which single collisions occur between the particles of the beam and the molecules of the gas. The radiation of the fast excited particles is recorded from a certain volume in the beam located at a definite distance from the entrance slit of the collision chamber, the absolute intensity of this radiation is determined, and the excitation cross section of the fast particle for this process is calculated. It is easy to show that this method can yield correct values of the excitation cross sections if the time of flight of the particles in the beam from the entrance slit to the region of observation is considerably greater than the lifetime of the excited states formed, i.e., under conditions of equilibrium between the production and radiation processes of the excited particles. If these times are comparable in magnitude, then such an equilibrium does not exist and the radiation intensity measured in the experiment is a function of the distance from the entrance slit to the region of observation. To determine the true values of excitation cross sections in this case, it is necessary to know the lifetimes of the excited particles. We will demonstrate this requirement by deriving a formula for the excitation cross section of fast particles.

Let a beam of ions with velocity v cm/sec and a density n_0^+ particles/cm^3 enter a gaseous target containing N atoms/cm^3. Interaction of the ions with the target atoms will result in appearance of both fast (incident) and slow (target) excited particles. Let us consider the change in the composition of the fast particle beam with time. Let n_j be the number of fast particles in state j at a certain moment of time. We neglect the change of velocity of the particles due to inelastic processes, i.e., we assume that the velocity of the secondary fast particles is equal to the velocity of the ions entering the collision chamber. Then the change of n_j with time can be described by a system of first order differential equations:

$$\frac{dn_j}{dt} = \sum_k n_k \sigma_{kj} N v + \sum_{k>j} n_k \gamma_{kj} - n_j \Gamma_j - n_j N v \sum_k \sigma_{jk}. \quad (1)$$

Here σ_{kj} is the cross section for the transition of the fast particle from the state k to the state j (k, j = 1, 2, . . . , k ≠ j); γ_{kj} is the probability of a spontaneous transition of the excited particle from the level k to the level j with radiation of the spectral line λ_{kj} corresponding to the transition; Γ_j is the total transition probability from the level j to all low-lying levels ($\Gamma_j = \sum_{i<j} \gamma_{ji}$). $\Gamma_j = 0$ for a stable state.

In Eq. (1) the first term $\sum_k n_k \sigma_{kj} N v$ takes into account the formation of particles n_j in collisions of all other particles n_k with gas atoms; the second term $\sum_{k>j} n_k \gamma_{kj}$ determines the population of the state j by radiative transitions from above (k > j). The remaining two terms describe the depopulation of the state j by spontaneous transitions to lower levels ($-n_j \Gamma_j$) and as the result of colli-

sions with gas atoms ($-n_j Nv\sum_k \sigma_{jk}$).

Let us find an approximate solution of Eq. (1) for the case of very low gas pressures when we can assume that mainly single collisions occur between the ions of the beam and the atoms of the target gas. For this purpose we will first separate in Eq. (1) the term determining the formation of n_j from primary ions n_0^+ and, using the law of conservation of number of particles $\sum_k n_k = n_0^+$, rewrite (1) in the form

$$\frac{dn_j}{dt} = n_0^+ Nv\sigma_{+j} + \sum_k n_k Nv(\sigma_{kj} - \sigma_{+j}) + \sum_{k>j} n_k \gamma_{kj} - n_j\Gamma_j - n_j Nv\left(\sigma_{+j} + \sum_k \sigma_{jk}\right). \quad (2)$$

Here σ_{+j} is the cross section for production of particles n_j in collisions of primary ions with gas atoms (hereafter we will denote this cross section simply as σ_j). We wish to find the solution of Eq. (2) in the region of single collisions, i.e., a solution proportional to the first power of N. Therefore we omit in (2) the terms proportional to N and consider the system of equations

$$\frac{dn_j}{dt} = n_0^+\sigma_j Nv + \sum_{k>j} n_k\gamma_{kj} - n_j\Gamma_j. \quad (3)$$

This system of equations describes the production of particles n_j by collisions of primary ions with gas atoms and by cascade transitions from levels higher than j, and the decay of particles n_j by spontaneous transition to all lower-lying levels. Neglecting in the first approximation the cascade transitions, we obtain

$$n_j = \frac{n_0^+\sigma_j Nv}{\Gamma_j}[1 - \exp(-\Gamma_j t)]. \quad (4)$$

Experimentally, we measure the number of photons q_{ji} corresponding to the transition $j \to i$ ($i < j$) radiated during the time of flight Δt of the fast particle in the region of observation. This quantity is determined by the relation $q_{ji} = \int_t^{t+\Delta t} n_j\gamma_{ji}dt$, where γ_{ji} is the probability of the transition $j \to i$. From this and from formula (4),

$$q_{ji} = \frac{n_0^+ Nv\sigma_j\gamma_{ji}\Delta t}{\Gamma_j}\left\{1 - \frac{[1 - \exp(-\Gamma_j\Delta t)]\exp(-\Gamma_j t)}{\Gamma_j\Delta t}\right\}, \quad (5)$$

or

$$q_\lambda = \sigma_\lambda n_0^+ N\Delta l\left\{1 - \frac{L_\lambda}{\Delta l}\left[1 - \exp\left(-\frac{\Delta l}{L_\lambda}\right)\right]\exp\left(-\frac{l}{L_\lambda}\right)\right\}, \quad (6)$$

where $\sigma_\lambda = \sigma_j\gamma_{ji}/\Gamma_j$ is the cross section for excitation of the spectral line λ corresponding to the transition $j \to i$ in the excited particle; $L_\lambda = v/\Gamma_j = v\tau_j$ is the characteristic length; τ_j is the lifetime of the state j; $l = vt$ is the distance from the entrance slit to the region of observation; $\Delta l = v\Delta t$ is the length of the beam from which the radiation of the excited particles is recorded; $q_\lambda = q_{ji}$.

Thus, if the experimental conditions are such that equilibrium is observed between the production and radiation of the excited fast particles, σ_λ is determined from the simple expression $q_\lambda = \sigma_\lambda n_0^+ N\Delta l$. This follows directly from formula (5) or (6) for $\Gamma_j t = t/\tau_j \gg 1$ (or $l \gg L_\lambda$). In the case when l is comparable with L_λ, the cross section σ_λ must be determined from the more general expression (6) which takes into account the lifetime of the excited fast particles.

In the present article we present a method which allows measurement of the lifetime of the excited states of fast particles under the same physical conditions in which the excitation cross sections of these particles are measured. We present experimental measurements, using this method, of the lifetime of hydrogen atoms excited to levels with principal quantum numbers n = 3, 4, 5.

2. METHOD OF MEASURING LIFETIMES OF EXCITED STATES OF FAST PARTICLES

1. Up to the present time the only method of measuring lifetimes of excited states of fast particles has been Wien's method, which was described in detail in 1927.[1] In this method the lifetime is determined from the attenuation of the luminosity of a beam of excited atoms in a vacuum. Wien's experiments on measurement of the lifetimes of excited hydrogen atoms have been the subject of serious criticism.[2,3] Nonuniform velocity composition of the excited hydrogen atom beam, indirect determination of the velocity of these atoms by the Doppler effect, the photographic method of measuring the radiation intensity, and insufficiently sharp drop in gas pressure at the exit aperture of the beam channel—all of these factors cast doubt on the accuracy of the results obtained by Wien.

It seems to us that Wien's method will not give accurate results even when used with present-day experimental techniques (monoenergetic beams of excited atoms, fast pumps, and photoelectric measurement of the radiation). In measuring the lifetime of an excited state j by this method, it is assumed that the state j in a vacuum is subject only to depopulation by spontaneous emission of light.

As a matter of fact, since the beam entering the vacuum from the collision chamber contains atoms not only in the state j but also in other excited states, as well as ions, another process also occurs—population of the state j: near the exit slit this results from excitation in collisions of ions and neutral atoms with molecules of the gas flowing through the slit (a very sharp pressure drop is hardly possible); and far from the exit slit, population of state j results from cascade transitions from higher levels. It is difficult to compute how much this process distorts the true attenuation curve for the luminescence of the excited atoms in the vacuum, and therefore lifetime measurements by Wien's method will always contain an indeterminate systematic error.

In the method which we propose, a monoenergetic beam of ions or atoms of a definite kind enters a gaseous target in which both production and radiative decay of fast excited atoms occur, and if no equilibrium exists between these processes, the intensity of the radiation from the excited atoms increases with distance from the entrance slit of the collision chamber. In this case, if we assume that cascade transitions do not play an important role in population of the state j, under conditions of single collisions between beam particles and gas atoms the number of photons q_λ emitted by a section of the beam of length Δl located at a distance l from the entrance slit is given by Eq. (6). The value of Δl is usually of the order of the spectrograph slit width, so that in many cases $\Delta l \ll L_\lambda$, and in that case

$$q_\lambda = \sigma_\lambda n_0^+ N \Delta l [1 - \exp(-l/L_\lambda)]. \quad (7)$$

This formula can already be used to determine the lifetime, by measuring the intensity of the line λ for several values of l. But since the pressure in the collision chamber and, because of the beam broadening, the effective length Δl from which light is collected can both change with l, direct application of this formula will not give accurate results. Furthermore, it is necessary to keep in mind that a pressure differential exists at the collision chamber entrance slit. To avoid the influence of all these effects on the accuracy of the measurement, we propose to utilize the fact that on passage of the ion or atom beam through the gas, excitation of gas atoms occurs along with the excitation of the fast particles. Under conditions of single collisions, the number of photons emitted by slow excited gas atoms (we can assume that the radiation of an excited gas atom occurs at the place where it is formed) is given by

$$q_\nu = \sigma_\nu n_0^+ N \Delta l, \quad (8)$$

where σ_ν is the excitation cross section of the spectral line characteristic of the gas atom, taking into account cascade transitions.

By measuring the intensity of the line emitted by the fast excited atom and relating it to the intensity of the line emitted by the excited gas atom, we exclude the effect of change of pressure and effective length Δl on the accuracy of the lifetime determination. Thus,

$$\frac{q_\lambda}{q_\nu} = \frac{\sigma_\lambda}{\sigma_\nu}\left[1 - \exp\left(-\frac{l}{L_\lambda}\right)\right],$$

or

$$\frac{J_\lambda}{J_\nu} = \frac{a_\lambda \sigma_\lambda}{a_\nu \sigma_\nu}\left[1 - \exp\left(-\frac{l}{L_\lambda}\right)\right], \quad (9)$$

where J_λ and J_ν are the photomultiplier anode currents in an optical-electrical detection system due to the line being studied and the comparison line, a_λ and a_ν are the transmission coefficients of this apparatus for the corresponding spectral lines. Using the designation

$$\frac{J_\lambda}{J_\nu} = y, \qquad a_\lambda \sigma_\lambda / a_\nu \sigma_\nu = b,$$

we can write

$$y = b[1 - \exp(-l/L_\lambda)]. \quad (10)$$

There is one further factor which must be considered. Formula (10) involves the quantity l which up to this time we have taken to be the distance from the ion beam entrance slit to the section Δl whose radiation is collected by the spectrograph. As a matter of fact, l is some effective distance $l' + \delta l$, where l' is the true geometrical distance from the entrance slit to the observation volume and $\delta l'$ is a correction arising from the pressure drop of the gas at the collision chamber entrance slit. Since the quantity δl enters into the argument of the exponential, it can have an important effect on the accuracy of lifetime measurements using formula (10). However, we can avoid this effect if we assume that it is the same for different values of l'.

The lifetime is then determined in the following way. Having measured the ratio of the intensity of the line being studied to the intensity of the comparison line along the path through the collision chamber, we construct a curve $y = f(l')$. As an initial point we can choose any point on this curve. Let this point be y_1 corresponding to l'_1. Taking two other points y_2 and y_3 corresponding to l'_2 and l'_3, so that $l'_3 - l'_2 = l'_2 - l'_1 = S$, we set up a

system of three equations:

$$y_1 = b\left[1-\exp\left(-\frac{l_1'+\delta l}{L_\lambda}\right)\right],$$

$$y_2 = b\left[1-\exp\left(-\frac{l_2'+\delta l}{L_\lambda}\right)\right]$$

$$= b\left[1-\exp\left(-\frac{l_1'+\delta l+S}{L_\lambda}\right)\right],$$

$$y_3 = b\left[1-\exp\left(-\frac{l_3'+\delta l}{L_\lambda}\right)\right]$$

$$= b\left[1-\exp\left(-\frac{l_1'+\delta l+2S}{L_\lambda}\right)\right],$$

from which we find

$$\exp(-S/L_\lambda) = (y_3-y_2)/(y_2-y_1).$$

From this the lifetime of the fast particle excited state is obtained as

$$\tau_j = S\left[v\ln\frac{y_2-y_1}{y_3-y_2}\right]^{-1}. \qquad (11)$$

Choosing different initial points l_1' and different values of S, we analyze the entire experimental curve in this way and determine the mean value of τ_j.

2. Let us discuss now the conditions under which we will have the greatest accuracy for measurement of fast particle excited states by the method described above. One of the conditions for the correct application of formula (10) to lifetime determination is obvious: the role of cascade transitions in population of the level studied must be small in comparison with direct excitation of this level in collisions. Returning to Eq. (3), we substitute in it the solution (4) for particles n_k. We obtain a system of equations

$$\frac{dn_j}{dt} = n_0^+\sigma_j Nv - n_j\Gamma_j + \sum_{k>j}\frac{n_0^+\sigma_k Nv\gamma_{kj}}{\Gamma_k}[1-\exp(-\Gamma_k t)], \qquad (12)$$

whose solution, expressed in terms of the number of photons q_λ, has for the case $\Gamma\Delta t \ll 1$ ($\Delta l \ll L$) the following form:

$$q_\lambda = \sigma_\lambda n_0^+ N\Delta l\left\{(1-e^{-\Gamma_j t}) + \sum_{k>j}\frac{\sigma_k\gamma_{kj}}{\sigma_j\Gamma_k}\left[1-\frac{\Gamma_j e^{-\Gamma_k t}-\Gamma_k e^{-\Gamma_j t}}{\Gamma_j-\Gamma_k}\right]\right\}. \qquad (13)$$

This solution takes into account that population of the level j occurs not only by the direct excitation process in collision of a fast ion with a gas atom (σ_j), but also by cascade transitions from higher levels k. The levels k are populated only by collisions (σ_k). It follows from (13) that, when $\Gamma t \gg 1$ ($l \gg L$), the greatest contribution to the measured intensity of the line λ is due to cascade transitions. In this case

$$q_\lambda = n_0^+ N\Delta l\left[\sigma_\lambda + \sum_{k>j}\sigma_k\frac{\gamma_{kj}\gamma_{ji}}{\Gamma_k\Gamma_j}\right].$$

Consequently in order to reduce the error from cascade transitions in lifetime measurements by the method proposed above, it is necessary to choose the length of the collision chamber as small as possible (of the order of or less than L_λ). If this requirement is not satisfied with sufficient accuracy, the value of lifetime determined from formula (10) will be larger than the true value.

Another systematic error in the method results from the following fact. Solutions (5) and (13) do not take into account that population of the level j can occur also by secondary collisions of fast excited particles n_j with gas atoms. This implies that in the equation system (2)

$$\Gamma_j \gg Nv\left(\sigma_j + \sum_k \sigma_{jk}\right). \qquad (14)$$

However, in certain cases, particularly in measuring the lifetimes of long-lived levels, it can turn out that these quantities are of the same order but

$$\left[\Gamma_j + Nv\left(\sigma_j + \sum_k \sigma_{jk}\right)\right]t \ll 1,$$

and a linear dependence of the intensity of the line studied on the gas pressure in the collision chamber is observed. It can be shown that we are then measuring not the true decay probability but some effective probability equal to

$$\Gamma_j + Nv\left(\sigma_j + \sum_k \sigma_{jk}\right).$$

Thus, for the correct application of the proposed method, it is necessary that condition (14) be satisfied. This means that the gas pressure in the collision chamber must be such that the experimentally measured lifetimes do not depend on gas pressure and on the velocity of the fast particles.

Analysis of formula (11) shows that the minimum random error in the lifetime measurements is obtained when $S \sim L_\lambda$ or $l \sim 2L_\lambda$.

3. MEASUREMENT OF LIFETIMES OF EXCITED STATES OF THE HYDROGEN ATOM

Quantum mechanics indicates[4] that the lifetimes of excited states of the hydrogen atom with a given principal quantum number n depend strongly on the population probabilities of the fine

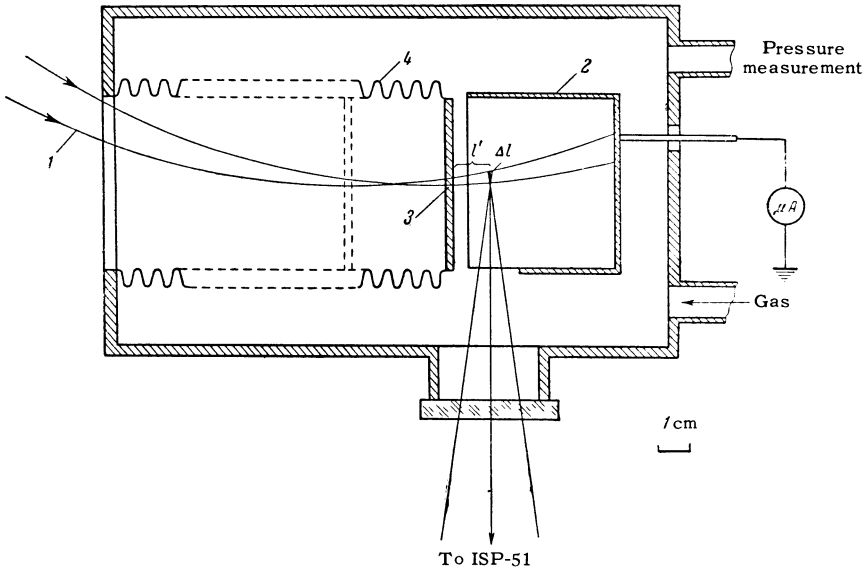

FIG. 1

structure sublevels, which in turn are determined by the excitation conditions. In the absence of external fields, the least lifetime of states with the same n belongs to the p-state; for a fixed azimuthal quantum number the lifetime varies as n^3. If the excitation conditions are such that population of the fine structure sublevels occurs according to the statistical weights of these sublevels, then quantum mechanics predicts the existence in this case of mean lifetimes which increase with n as $n^{4.5}$.

As we have already mentioned, experimental determinations of lifetimes of excited states of the hydrogen atom have been carried out by Wien.[1] Wien's results contradict quantum mechanics: he obtained the same values of lifetime for levels with n = 3, 4, and 5. Wien's experiments were repeated by Atkinson[2] and Von Ardenne,[3] but the contradiction remains. Furthermore, Von Ardenne, by measuring the attenuation of the H_β line in vacuum, found that the excited hydrogen atom lifetimes are in agreement with classical electrodynamics. Thus, up to the present time there are no accurate experimental data on excited hydrogen atom lifetimes which would correspond to quantum mechanical calculations.

On the other hand, the problem of the lifetimes of the excited states of the hydrogen atom has currently acquired a practical interest in connection with the problem of injecting excited hydrogen atoms into magnetic traps. Therefore the measurement of these times by the new method described above was of some interest.

The experimental apparatus in which our measurements of excited hydrogen atom lifetimes were made has been described in detail in our previous paper.[5] A beam of hydrogen ions from a source was broken down into its components in a mass spectrometer with 180° deflection of the beam. H_2^+ ions of energy 10—30 keV were directed into a collision chamber filled with helium.

A drawing of the collision chamber is shown in Fig. 1. The collision chamber entrance slit 3, mounted on a bellows 4, could be moved along the ion beam 1. The displacement of the entrance slit corresponded to a variation from 0—30 mm of the distance l' from the slit to the section of the beam whose radiation was detected by the spectrograph (Δl = 0.2 mm). The collision chamber was in the magnetic field of the mass spectrometer. The H_2^+ ion current was measured by the collector 2.

We measured the lifetimes of hydrogen atoms excited to levels with principal quantum numbers n = 3, 4, and 5 in the dissociation of H_2^+ ions in helium. The first three lines of the Balmer series, H_α, H_β, H_γ, were used for these measurements; the He I line λ = 5876 Å (transition $3^3D \rightarrow 2^3P$) was selected as the comparison line. For a given H_2^+ ion energy, we measured the ratio of the intensities of the H_α, H_β, and H_γ lines to the intensity of the He I line λ = 5876 Å at different distances l' from the entrance slit and plotted the variation of the ratio J_{H^*}/J_{He^*} with l'.

Figure 2 shows experimental curves whose ordinates have been normalized by coefficients b so that the behavior of these curves is due only to the difference in lifetimes of the corresponding excitation levels. Curves of a similar type were obtained for different H_2^+ ion energies and different helium pressures in the collision chamber, in order to

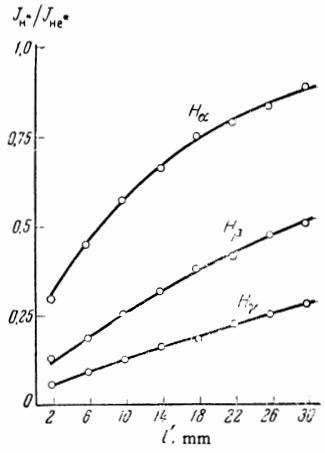

FIG. 2

verify the dependence of the measured lifetimes on these parameters. The range over which the pressure was varied corresponded to the region of linear dependence on pressure both for the Balmer lines and for the comparison lines. The experimental curves were analyzed according to formula (11), and thus the average value of the lifetime was computed for each of the three excited hydrogen atom levels studied. The results of the measurements are given in the table; listed in the same table for comparison are the values obtained by Wien[1] and Atkinson[2] and the theoretical values corresponding to a statistical population of the fine structure sublevels.[4]

As follows from the table presented, the lifetime values obtained by the present authors are in good agreement with the quantum mechanical calculations. For the line H_α the experimental lifetime is somewhat larger than the theoretical value. This is due to the fact that the experimental conditions for the H_α line were not so favorable as for the H_β line. Calculations using the known transition probabilities in the hydrogen atom[4] and the measured excitation cross sections of the Balmer lines[5] enable us to evaluate by means of formula (13) the contribution of cascade transitions to the intensity of the lines studied. It turns out that this contribution is greatest for the line H_α: for $l' = 30$ mm it amounts to about 6%, which is sufficient to exaggerate the measured lifetime. Condition (14) is not sufficiently well fulfilled for the H_γ line, and therefore we observe some reduction of the mean lifetime in comparison with the theoretical value.

The agreement between the experimental and theoretical lifetime values calculated for the case of a statistical population of the fine structure sublevels, it seems to us, is due to the fact that in our experiments the hydrogen atom excitation occurred

Lifetimes τ of excited states of the hydrogen atom

Line	Principal quantum number n	τ, 10^{-8} sec			
		Present work	Theory [4]	Wein's data [1]	Atkinson's data [2]
H_α	3	1.25±0.1	1,02	1.85	1,80
H_β	4	3.4±0.15	3.35	1.85	3.2
H_γ	5	7,8±1.5	8,8	1.85	—

in a magnetic field. The magnetic field strength varies from 1000 to 1800 Oe, and since the velocities of the fast particles are $1 - 2 \times 10^{-8}$ cm/sec, we can consider that an equivalent electric field with an intensity of 1—3.6 kV/cm acts on the excited hydrogen atom formed, which leads to a Stark splitting of the levels. The critical electric field intensity at which the Stark splitting of the $n = 3, 4, 5$ levels becomes comparable with their total fine structure splitting is 400, 100 and 30 V/cm, respectively.[6] Consequently under our experimental conditions the Stark splitting was considerably larger than the fine structure splitting; in this case a mixing of states with different azimuthal quantum numbers occurs and the lifetimes of the levels correspond to a statistical population of the fine structure sublevels.

In conclusion we list the results of measuring the lifetimes of the 4^3S and 3^3P levels of the helium atom, which were carried out as a check on the method. Experimental[7] and theoretical[8] data exist for the lifetimes of these levels. The excited helium atoms were obtained by charge exchange of 10—20 keV He^+ ions in hydrogen. The ratio of intensities of the He I lines $\lambda = 4713$ Å ($4^3S \rightarrow 2^3P$) and $\lambda = 3889$ Å ($3^3P \rightarrow 2^3S$) to the intensity of the H_β line was measured for different distances l' from the collision chamber entrance slit. The lifetime was determined according to formula (11). The values obtained are in satisfactory agreement with the results of other experiments[7] and with theory[8]. In units of 10^{-8} sec, the lifetimes of the helium atom levels investigated are as follows: $\tau(4^3S) = 6.8 \pm 0.2$ (the present investigation), 6.75 ± 0.1[7], 6.4[8]; $\tau(3^3P) = 10 \pm 1$ (the present investigation), 11.5 ± 0.5[7], 9.7[8].

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