EXCITATION OF 3s, 3p, AND 3d STATES OF THE HYDROGEN ATOM UPON DISSOCIATION OF THE H *MOLECULE BY FAST* He *IONS*

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We describe the theory of the method and present experimental results of the measurement of the partial cross sections σ_{3s}, σ_{3p}, and σ_{3d} for the excitation of hydrogen atoms upon dissociation of the H_2 molecule by He^+ ions with energies 10—30 keV.

It is of interest to investigate the dependence of the cross sections for the excitation of hydrogen atoms on the azimuthal quantum number l, in order to clarify the character of the population of the fine-structure sublevels of the hydrogen atom during the process of its excitation.

In this paper we describe a method which makes it possible to solve this problem to some degree. We present below the theory of a method and experimental results for the measurement of the partial cross sections σ_{3s}, σ_{3p}, and σ_{3d} for the excitation of the hydrogen atoms upon dissociation of a H_2 molecule by He^+ ions with energies 10—30 keV.

1. THEORY OF THE METHOD

Since the level of the hydrogen atom with $n = 3$ consists of three fine-structure sublevels 3s, 3p, and 3d, it is possible in principle, by writing three independent equations relating certain experimental quantities and the excitation cross sections σ_{3s}, σ_{3p}, and σ_{3d} of the hydrogen atom, and by solving these equations, to determine the absolute values of these cross sections. The experimental quantities can be the following:

A. Intensity of the H_α line in the case when there are no electric and magnetic fields in the collision chamber (the zero-field case). The cross section for the excitation cross section σ_λ of the H_α in a zero field is expressed in terms of the partial cross sections by the relation

$$\sigma_\lambda = \sigma_{3s} + \frac{\gamma_{3p}^{2s}}{\Gamma_{3p}}\sigma_{3p} + \sigma_{3d} = \sigma_{3s} + 0.1183\sigma_{3p} + \sigma_{3d}, \quad (1)$$

in which γ_{3p}^{2S} is the probability of the $3p \to 2s$ transition, and Γ_{3p} is the total probability of the decay of the 3p level in the hydrogen atom.

B. The ratio of the intensity I_{H_α} of the H_α line, excited in the gas in a strong electric field, to its intensity J_{H_α} in a zero field

$$A = I_{H_\alpha}/J_{H_\alpha} = \varphi_1(\sigma_{3s}, \sigma_{3p}, \sigma_{3d}). \quad (2)$$

C. The degree of polarization of the radiation of the H_α line in the same field, or more accurately the ratio of the intensity I_z of the H_α radiation polarized along the direction of the electric field, to the intensity I_x of the radiation polarized transverse to the field direction,

$$g = I_z/I_x = \varphi_2(\sigma_{3s}, \sigma_{3p}, \sigma_{3d}). \quad (3)$$

The form of the functions φ_1 and φ_2 can be obtained with the aid of quantum mechanics of the hydrogen atom. Following Schrödinger [1], we have

$$I_z = K(729f_{110} + 2304f_{101} + 1682f_{200}), \quad (4)$$

$$I_x = I_y = K(441f_{110} + 1952f_{101} + 18f_{200} + 2304f_{002}). \quad (5)$$

Here K is a numerical coefficient, which can be determined from the correspondence principle, $f_{n_1, n_2, m}$ is the density matrix which determines the radiation of the atom in a strong electric field, n_1 and n_2 are the parabolic quantum numbers, and m is the magnetic quantum number. For zero field we have

$$J_z = J_x = J_y = K(75f_{3s} + 800f_{3p} + 3840f_{3d}). \quad (6)$$

Here f_{3l} is the density matrix, which determines the radiation of the atom in the zero field, and l is the azimuthal quantum number. In the derivation of these relations it is assumed that the population over the magnetic quantum numbers is equally probable in the process of excitation of the hydrogen atom in the zero field, i.e., the degree of polarization of the H_α line in zero field is zero.

The density matrix f, which determines the radiation of the atom, can be expressed by the equation $f = \rho/\Gamma$, where Γ is the probability of radiative decay, in terms of the density matrix ρ, which characterizes the excitation of the atom. By determining the coupling coefficients between the wave functions of the hydrogen atom for the level with n = 3 in parabolic coordinates and the wave functions in spherical coordinates, and by using the definition of the density matrix, we obtain, omitting the intermediate steps

$$\begin{aligned} f_{002} &= \rho_{3d}/\Gamma_{002}, \\ f_{101} &= (\rho_{3p}+\rho_{3d})/2\Gamma_{101}, \\ f_{110} &= (\rho_{3s}+2\rho_{3d})/3\Gamma_{110}, \\ f_{200} &= ({}^1/_3\rho_{3s}+{}^1/_2\rho_{3p}+{}^1/_6\rho_{3d})/\Gamma_{200}. \end{aligned} \tag{7}$$

Here $\Gamma_{n_1, n_2, m}$ is the probability of the decay of the Stark sublevels in the hydrogen atom, and the ρ_{3l} are related with the partial excitation cross sections by the following equations:

$$\sigma_{3s}\sim\rho_{3s}, \quad \sigma_{3p}\sim 3\rho_{3p}, \quad \sigma_{3d}\sim 5\rho_{3d}. \tag{8}$$

By calculating $\Gamma_{n_1, n_2, m}$ and Γ_{3l} with the aid of the quantum mechanics of the hydrogen atom[2] and substituting (7) and (8) in (4), (5), and (6), and then further in (2) and (3), we obtain

$$A=\frac{I_{H_\alpha}}{J_{H_\alpha}}=\frac{I_z+2I_x}{J_z+2J_x}=\frac{0{,}4825\sigma_{3s}+0{,}3029\sigma_{3p}+0{,}6852\sigma_{3d}}{\sigma_\lambda}, \tag{9}$$

$$g=\frac{I_z}{I_x}=\frac{0.2969\sigma_{3s}+0.1577\sigma_{3p}+0.1258\sigma_{3d}}{0.0928\sigma_{3s}+0.0726\sigma_{3p}+0.2797\sigma_{3d}}. \tag{10}$$

From these equations and Eq. (1) we obtain finally

$$\begin{aligned} \frac{\sigma_{3s}}{\sigma_\lambda} &= 1.046+\frac{A}{g+2}(-4.270+1.180g), \\ \frac{\sigma_{3p}}{\sigma_\lambda} &= -2.133+\frac{A}{g+2}(5.114+5.587g), \\ \frac{\sigma_{3d}}{\sigma_\lambda} &= 0.206+\frac{A}{g+2}(3.665-1.841g). \end{aligned} \tag{11}$$

From this we can obtain the ratio of the total cross section for the excitation of the level with n = 3 to the excitation cross section of the H_α line

$$\frac{\sigma_3}{\sigma_\lambda}=-0.881+\frac{A}{g+2}(4.509+4.926g). \tag{12}$$

Thus, by measuring the ratio of the intensity of the H_α line in a strong electric field to its intensity in a zero field, and by measuring the polarization factor g of the H_α radiation in the electric field, we can determine the relative magnitudes of the cross sections for the excitation of the fine-structure sublevels of the hydrogen atom for n = 3, and by determining the absolute value of the excitation cross section σ_λ of the H_α line in a zero field, using the ordinary procedure of comparing its intensity with the intensity of an absolutely black body, we can obtain the absolute values of the cross sections σ_{3s}, σ_{3p}, and σ_{3d}.

2. EXPERIMENT AND DISCUSSION OF RESULTS

The choice of the process of dissociation of the hydrogen molecule with formation of excited hydrogen atoms was based on the assumption, on which the theory of the method is based, that the H_α radiation in the absence of a field is not polarized. Since in this case there is no preferred direction in space with respect to the emission of the produced excited hydrogen atoms, it was expected that such an assumption is valid. Experiment has shown in fact that the degree of polarization of the H_α line in a zero field is equal to zero in the entire investigated energy interval.

The setup used for the measurements is described in our paper[3]. The beam of He^+ ions entered a collision chamber filled with hydrogen to pressures $5\times10^{-4}-1\times10^{-3}$ mm Hg. The observation of the H_α emission was at angle of 90° relative to the direction of motion of the ion beam. With the aid of a parallel-plate capacitor it was possible to produce in the collision chamber an electric field perpendicular to the motion of the ion beam and to the direction of the observation of the H_α line. The radiation was recorded with a three-prism ISP-51 spectrograph and an FÉU-38 photomultiplier. The separation of the radiation polarized parallel or perpendicular to the electric field direction was with the aid of a polaroid made of a "gerapatite" film. To eliminate the unequal sensitivity of the prism monochromator to light with different polarization directions, a quarter-wave plate was placed ahead of the entrance to the monochromator and transformed the linearly-polarized radiation components into light with circular polarization. The entire optical system was adjusted by passing unpolarized light through it.

We investigated the dependence of the total intensity of the H_α line excited by single collisions between the He^+ ions and the H_2 molecules, on

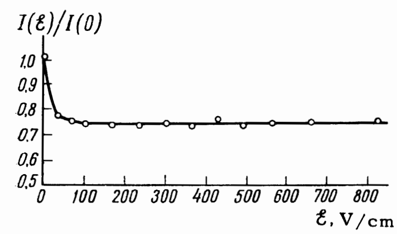

FIG. 1. Intensity of the H_α line vs. electric field intensity ℰ in the collision chamber (collisions between He^+ and H_2; He^+ ion energy is E = 20 keV).

	A	g	P_{H_α}	σ_{3s}/σ_3	σ_{3p}/σ_3	σ_{3d}/σ_3
Experimental results of $He^+ + H_2 \rightarrow H_\alpha$	0.81±3%	0.74±5%	−0.15	0.026± ±100%	0.400± ±10%	0.574± ±10%
Statistical population of fine-structure sublevels	1	1	0	0.007	0.633	0.360
Dynamic population of the fine-structure sublevels	0.76	0.82	−0.10	0.111	0.333	0.556

the intensity of the electric field in the collision chamber. A typical plot is shown in Fig. 1. When the electric field is applied, a decrease is observed in the intensity of the H_α radiation, owing to the redistribution of the excitation among the sublevels of the fine structure under the influence of the field. When the field intensity exceeds a value on the order of 100 V/cm, the intensity of the H_α line remains unchanged. Measurements of the z and x components of the H_α lines were made at an electric field intensity $\mathscr{E} = 500-800$ V/cm. On the basis of Fig. 1, such a field can be regarded as "strong" enough to justify the use the Stark wave functions for the hydrogen atom in the theory of the method.

The values of A and g were determined in the incident He^+ ion energy range from 10 to 30 keV. It turned out that these quantities do not depend on the ion energy and their average values were $A = 0.81$ and $g = 0.74$. From these data we calculated with the aid of (11) and (12) the relative partial cross sections σ_{3s}/σ_3, σ_{3p}/σ_3, and σ_{3d}/σ_3; their values are listed in the table, while Fig. 2 shows plots of the absolute cross sections σ_{3s}, σ_{3p}, and σ_{3d} and of the total level-excitation cross section σ_3 against E (the energy of the He^+ ions). The error in the measurements of the absolute intensity of the H_α line is 20%.

Besides the experimental results, the table also lists the values of A, g, the degree of polarization P_{H_α} of the H_α line in the electric field, and the

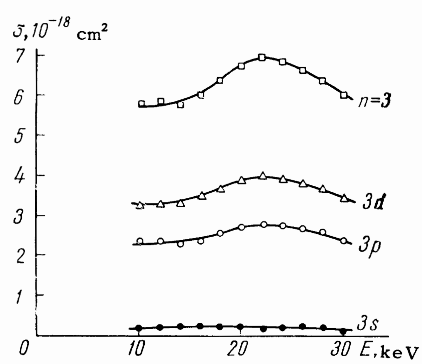

FIG. 2. Cross sections for the excitation of the fine-structure sublevels of the hydrogen atom for n = 3 (collisions of He^+ with H_2; E–energy of He^+ ions).

relative excitation cross sections of the sublevels 3s, 3p, and 3d for the two cases considered in the quantum mechanics of the hydrogen atom [2]: a) when excitation and radiative decay establish, on the average, a distribution of the excited hydrogen atoms over the fine-structure sublevels corresponding to the statistical weight of these sublevels (statistical population of the fine-structure sublevels), and b) when the excitation is in accordance with the statistical weights of the fine-structure sublevels, and the established distribution of the excited atoms over the states with different quantum numbers l is determined by the lifetimes of these states (dynamic population of the fine-structure sublevels). In the former case all the f, and in the latter case all the ρ are the same for different azimuthal quantum numbers, and the values listed in the table can be readily obtained with the aid of (4)–(12). It follows from the table that in the process $He^+ + H_2 \rightarrow H^*$ investigated by us, none of the cases considered by the theory is realized.

We verified the proposed method by measuring with the aid of a vacuum monochromator, with diffraction grating, the ratio of the intensity I_{L_β} of the L_β line in a strong electric field to its intensity J_{L_β} in a zero field (the L_β line, $\lambda = 1026$ Å, occurs in the 3p – 1s transition in the hydrogen atom). An equation for this ratio can be obtained with the aid of quantum mechanics on the basis of (7) and (8), and in simpler fashion from the condition that the total number of quanta of the H_α and L_β emissions should be proportional to the excitation cross section of the level σ_3 and does not depend on the presence of a field in the collision chamber. Then we get from (1) and (9)

$$\frac{I_{L_\beta}}{J_{L_\beta}} = \frac{0.5175\sigma_{3s} + 0.6971\sigma_{3p} + 0.3148\sigma_{3d}}{0.8817\sigma_{3p}}. \quad (13)$$

Using the measured cross sections from the table and Eq. (13), we get $I_{L_\beta}/J_{L_\beta} = 1.34$, which is in good agreement with experiment: the intensity of the L_β line increases when an electric field is applied to the collision chamber. From the equation $g_{L_\beta} = I_z/I_x = f_{200}/f_{101}$, we can calculate also the degree of polarization of the L_β line in the electric field. It is found to be −0.05.

In conclusion we note that the proposed method and the presented experimental results can be used to determine the absolute intensity of the L_β line. Knowing the value of the cross section for the excitation of the 3p state of the hydrogen atom, we get the excitation cross section of the L_β line from the equation

$$\sigma_{L_\beta} = \frac{\gamma_{3p}^{1s}}{\Gamma_{3p}} \sigma_{3p} = 0{,}8817 \sigma_{3p}, \tag{14}$$

in which γ_{3p}^{1S} is the probability of the $3p \rightarrow 1s$ transition.

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