CHARGE EXCHANGE OF PROTONS IN INERT GASES INVOLVING THE FORMATION OF FAST HYDROGEN ATOMS IN THE 2s AND 2p STATES

E. P. ANDREEV, V. A. ANKUDINOV, and S. V. BOBASHEV

A. F. Ioffe Physico-technical Institute, Academy of Sciences, U.S.S.R.

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The cross sections $\sigma(2s)$ and $\sigma(qp)$ for the excitation of the 2s and 2p states of hydrogen atoms, produced in the charge exchange of 10–40 keV protons in helium, neon, argon, krypton, and xenon, were determined by measuring the intensity of the first line of the Lyman series. The metastable 2s state of the hydrogen atom was de-excited by establishing an electric field in the charge exchange chamber. The absolute intensity of the L_α line was determined from the photoionization current in nitric oxide. In the investigated range of proton energies, the cross section $\sigma(2s)$ was found to increase monotonically only in helium; in all other cases the energy dependence of $\sigma(2p)$ and $\sigma(2s)$ exhibited maxima. The maximum values of the cross sections increased as the atomic number of the target gas increased and reached $\sigma_m(2s) = 8.7 \times 10^{-17}$ cm^2 (Xe, E = 17 keV) and $\sigma_m(2p) = 1.2 \times 10^{-16}$ cm^2 (Xe, E = 11 keV). At proton energies E < 25 keV, the cross sections were $\sigma(2s) < \sigma(2p)$; for E > 25 keV, the ratio $\sigma(2s)/\sigma(2p) > 1$ and it increased as the atomic number of the target gas decreased. [For helium, $\sigma(2s)/\sigma(2p) = 4$ at E = 40 keV.]

I. INTRODUCTION

In the hydrogen atom, the level with the principal quantum number n = 2 is split into three sublevels: $2s^2S_{1/2}$, $2p^2P_{1/2}$, and $2p^2P_{3/2}$.[1] Of these, the 2s level is metastable, because the 2s–1s transition is forbidden by the selection rules for the electric dipole radiation. However, the lifetime τ of the 2s level is finite because a transition is possible to the ground level 1s by the emission of two photons. Quantum-mechanical calculations give 0.14 sec for the lifetime $\tau(2s)$.[1]

Fast hydrogen atoms formed by the charge exchange between protons and gas atoms are partly in excited states, including the 2s state. Some investigators[2-5] have determined the number of hydrogen atoms in the 2s state by a method based on the "mixing" of the 2s and 2p levels in an external electric field. Due to the Lamb shift, the $2s_{1/2}$ level of the hydrogen atom lies 0.035 cm^{-1} above the $2p_{1/2}$ level.[6] An external electric field induces Stark splitting, as a result of which the $2s_{1/2}$–$2p_{1/2}$ transition becomes possible. The hydrogen atom goes over from the $2p_{1/2}$ state to the ground state by emitting the first Lyman line L_α (1216 Å). Sellin[2] has shown that the lifetime of the $2s_{1/2}$ state in the transition $2s_{1/2}$–$2p_{1/2}$–$1s_{1/2}$ depends on the external electric field intensity and its minimum value, $\approx 3 \times 10^{-9}$ sec, is obtained at a field intensity ≈ 500 V/cm. In the cited investigations,[2-5] a beam of hydrogen atoms, formed by passing protons through a gas chamber, which included excited atoms, was admitted into an evacuated chamber and subjected to an electric field. The radiation of the L_α line, produced by the 2s–2p–1s transition, was recorded. The cross section for the excitation of the 2s state in the charge exchange of protons could be calculated from the measured absolute intensity of this radiation.

A disadvantage of this method was the fact that it was practically impossible to determine the total number of hydrogen atoms in the 2s state, formed in the collision chamber, since on the way from the collision chamber to the boundary of the region where an external electric field was applied, some metastable atoms were lost by processes which de-excited the 2s state (this state could be de-excited by stray fields, collisions with molecules of the residual gas, etc.). We employed a method which made it possible to overcome this difficulty.[1)]

An electric field, which de-excited the metastable state, was established in the collision chamber

[1)] A similar method was used independently by Jaecks, van Zyl, and Geballe.[7]

itself. When a beam of protons was passed through a gas target, hydrogen atoms in the 2s and 2p states were formed continuously by the charge exchange process. In the absence of an external electric field, the L_α radiation was solely due to the decay of the 2p state. When an external field was applied, the intensity of the L_α radiation increased as the field intensity was increased, indicating that this radiation was due not only to the 2p–1s transition but also to the 2s–2p–1s transition. At some value of the electric field intensity, corresponding to the minimum value of the lifetime of the 2s state, the intensity of the L_α radiation became constant. From the values of the intensity of this radiation in the presence and in the absence of a field, we could calculate the cross section for the excitation of the 2s state of the hydrogen atom.

Using this method, we investigated the charge exchange of protons in inert gases involving the formation of fast hydrogen atoms in the 2s and 2p states. We obtained the absolute values of the effective cross sections for the excitation of 2s and 2p states of the hydrogen atom by the processes

$$H^+ + He(Ne, Ar, Kr, Xe) \rightarrow H(2s, 2p) + He^+(Ne^+, Ar^+, Kr^+, Xe^+)$$

in the proton energy range 10–40 keV.

II. APPARATUS

1. The apparatus is shown schematically in Fig. 1. Part of the apparatus, including an ion source 2 and a mass spectrometer 1, was described earlier.[8,9] A monokinetic beam of protons was extracted, by means of a magnetic screen 3 and a system of electrostatic quadrupole lenses 4, from the mass spectrometer chamber into a collision chamber filled with a gas to a pressure at which single collisions between the beam particles and the gas atoms predominated. A parallel-plate capacitor 5 separated the proton beam from the neutral component. Over the whole path of the ion beam between the mass-spectrometer chamber and the collision chamber, the pressure did not exceed $(2–3) \times 10^{-6}$ mm Hg. In the collision chamber, we mounted: a parallel-plate capacitor 8, in whose field the metastable states of the hydrogen atom were de-excited, an extensible ion detector 7, and a trap 9 filled with liquid nitrogen.[2)] During the measurements, the constancy of the ion current was checked with another ion detector (not shown in Fig. 1), placed behind the "de-excitation" capacitor 8.

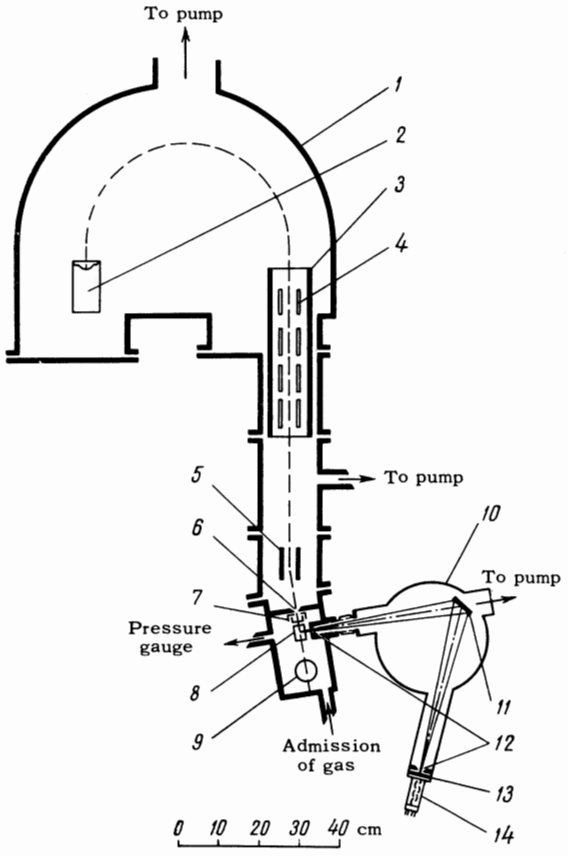

FIG. 1. Schematic diagram of the apparatus: 1) vacuum chamber of the mass spectrometer; 2) ion source; 3) magnetic screen; 4) electrodes of quadrupole lenses; 5) deflecting capacitor; 6) entry slit of the collision chamber; 7) extensible ion detector; 8) capacitor producing the de-exciting electric field; 9) trap; 10) vacuum monochromator; 11) bent diffraction grating; 12) entry and exit slits of the vacuum monochromator; 13) screen covered with sodium salicylate; 14) photomultiplier.

2. The L_α radiation, emitted by fast excited hydrogen atoms in a region in the beam at a distance of 35 mm from an entry slit of the collision chamber, reached a vacuum monochromator of the Seya–Namioka type.[10] In this monochromator, we used a replica of a bent diffraction grating 11 whose radius of curvature was 0.5 m, and which had 1200 lines/mm and was coated with a film of magnesium fluoride. The L_α radiation at the exit from the monochromator was detected with a photomultiplier FÉU-29 (14), which recorded the visible light excited by the L_α radiation in a thin film (≈ 2.5 mg/cm^2) of sodium salicylate, deposited on a screen 13.

III. MEASUREMENT METHOD

1. Relative measurements of the L_α radiation intensity. By observing the single-collision condi-

[2)] In the case of krypton and xenon, the trap was filled with a mixture of acetone and solid carbon dioxide.

tions, we measured the excitation functions of the 2s and 2p states of the hydrogen atom in the charge exchange of protons in inert gases, in the energy range 10–40 keV. All the measurements of the L_α radiation intensity, in relative units, were made with a vacuum monochromator[3] using a photoelectric recording system. The intensity of the L_α radiation, $I_{L_\alpha}(2p)$, corresponding to the 2p–1s transition, was measured in the absence of an external electric field. After the application of the field in the collision chamber, we measured the intensity of the L_α radiation due to the decay of the 2s and 2p sublevels. From these measurements, we determined that part of the L_α radiation intensity $I_{L_\alpha}(2s)$ which corresponded to the decay of the 2s state. The intensity of the electric field at which the 2s lifetime reached its minimum value, was determined experimentally. Figure 2 shows the dependence of the ratio $I_{L_\alpha}(2s)/I_{L_\alpha}(2p)$ on the external field intensity for the charge exchange of protons of 20 and 30 keV energies in argon. It is evident from Fig. 2 that the $I_{L_\alpha}(2s)/I_{L_\alpha}(2p) = f(\mathscr{E})$ curve reached saturation [minimum value of $\tau(2s)$] at a field intensity $\mathscr{E} \approx 500$ V/cm. All the measurements were carried out at an external field intensity $\mathscr{E} = 600$ V/cm.

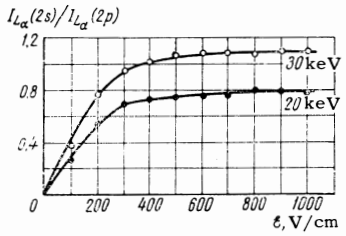

FIG. 2

In the present study, we investigated the fast excited hydrogen atoms. To measure correctly the intensity of light emitted by a fast particle, the time of flight of the particle from the entry slit of the collision chamber to the region of observation (for metastable atoms—from the front boundary of the de-exciting field to the region of observation) must be much greater than the lifetime of the generated fast excited particles.[11] The length of the plates of the "de-excitation" capacitor and the distance from the entry slit of the collision chamber to the region of observation were selected in such a way as to satisfy this condition.[4]

Later, in a separate experiment at a proton energy of 16 keV, we carried out comparative measurements of the L_α radiation intensities resulting from the excitation of the 2p state of the hydrogen atom by the charge exchange of protons in all the investigated gases. From these measurements, we determined the coefficients for the transformation from the excitation cross section of the L_α line in a given gas to the excitation cross sections in the other gases. This made it possible to restrict the absolute measurements of the L_α radiation intensity to only one process and one proton energy.

2. Absolute measurements of the L_α radiation flux. The absolute measurements of the L_α radiation intensity by means of a vacuum monochromator would have required preliminary calibration of the whole recording apparatus, i.e., the determination of the admittance coefficient of the vacuum monochromator, of the radiation detector sensitivity, and of the quantum yield of sodium salicylate. The complexity of such measurements forced us to abandon an attempt to calibrate the optical channel. Because of the high density of the ion current in our apparatus (up to 1 mA/cm^2 in the collision chamber), we found it possible to record the resultant L_α radiation with an ionization chamber. Figure 3 shows the attachment to our apparatus, used in the absolute measurements of the L_α radiation intensity.

An ionization chamber 8 with a lithium fluoride window 7, filled with nitric oxide NO to a pressure of 1–2 mm Hg, was attached directly to the collision chamber 1. The radiation emitted during the passage of the proton beam through the gas target reached the measuring region of the ionization chamber and photoionized the NO molecules. The photoionization current determined was a measure of the L_α intensity only if, in the spectral interval determined by the transmission limit of the LiF window (1050 Å) and the photoionization threshold of the NO molecule (1346 Å), no other radiation except the L_α line was excited in the collision chamber. Measurements using the vacuum monochromator showed that this condition was satisfied by the radiation generated in the process $H^+ + Ne \rightarrow L_\alpha$. The ionization chamber we constructed (Fig. 3) was fitted with two measuring electrodes

[3] The entry and exit slits of the monochromator were, respectively, 1 × 20 mm and 2 × 40 mm. In the case of krypton, the slits were narrowed in order to separate the 1216 Å (L_∞) and 1236 Å (Kr I) lines.

[4] In our case, the effective boundary of the de-exciting electric field was not less than 2 cm from the observation region.

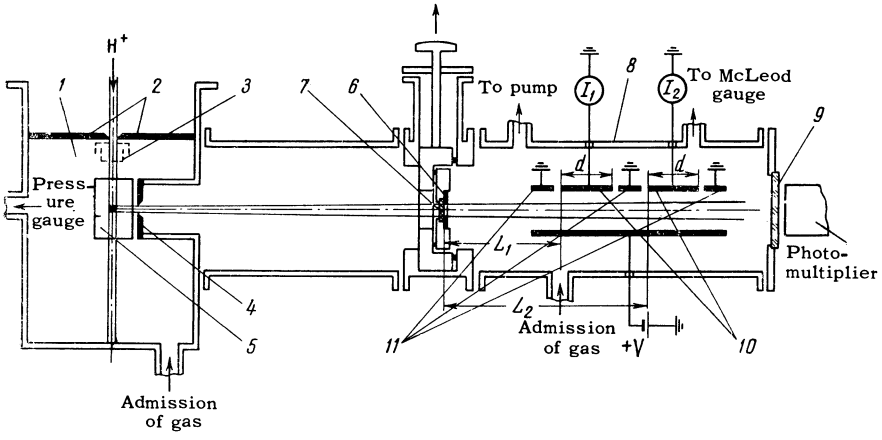

FIG. 3. Experimental arrangement used in the absolute measurement of the intensity of the L_α line: 1) collision chamber; 2) entry slit to the collision chamber; 3) extensible ion detector; 4) front diaphragm of the ionization-chamber collimator; 5) capacitor producing the de-exciting electric field; 6) back diaphragm of the ionization-chamber collimator; 7) LiF window; 8) ionization chamber; 9) screen covered with sodium salicylate; 10) measuring electrodes in the ionization chamber; 11) guard electrodes in the ionization chamber.

10 and could be used to measure the flux of quanta entering the chamber without a direct measurement of the gas pressure.

The flux of quanta, $I^0_{L_\alpha}$, entering the ionization chamber, and the photoionization current flowing to the measuring electrode, i_1, are related by[12]

$$I_{L_\alpha}{}^0\gamma = \frac{i_1/e}{\exp(-\mu L_1)[1-\exp(-\mu d)]}, \qquad (1)$$

where L_1 is the distance from the entry window of the ionization chamber to the front edge of the first measuring electrode; d is the length of the measuring electrode (25 mm); μ is the linear coefficient of absorption of the L_α radiation in nitric oxide at a given pressure; γ is the photoionization yield for the NO molecules; e is the electronic charge. A similar expression can be written down for the second measuring electrode. From these relationships, we can easily calculate μ:

$$\mu = \frac{\ln(i_1/i_2)}{L_2 - L_1}.$$

Here, i_2 is the photoionization current flowing to the second measuring electrode; L_2 is the distance from the entry window of the ionization chamber to the front edge of the second measuring electrode. The gas pressure in the ionization chamber does not appear explicitly in Eq. (1). Consequently, it was unnecessary to measure the NO pressure at the moment of carrying out the experiment. To check the results, we determined independently the coefficient μ by measuring the attenuation of the L_α radiation in the chamber. These two independent measurements gave results which agreed within the limits of the experimental error.

The purity of nitric oxide was checked by means of a mass spectrometer. Foreign impurities did not exceed 4%. Nitric oxide was admitted to the ionization chamber via a trap cooled by a mixture of acetone and solid carbon dioxide. The value of the quantity γ, equal to 0.83, was taken from Watanabe's paper.[13] The transmission coefficient of the LiF window was determined twice in separate experiments: before and after measurements in the ionization chamber. In our calculations, we used an average value, equal to 0.40 (the error was $\approx 3\%$).

The ionization chamber was used to measure the absolute intensity of the L_α radiation produced by the charge exchange of 16 keV protons in neon in the absence of an electric field. The measured flux of the L_α quanta, entering the ionization chamber, was 1.0×10^7 quanta/sec. (The light was recorded over a beam length $\Delta l = 0.32$ cm in a solid angle $\approx 2 \times 10^{-3}$ sr at a neon pressure of $\approx 1 \times 10^{-3}$ mm Hg in the collision chamber and a proton current density of 0.6 mA/cm^2.) This flux corresponded to a current of 1.8×10^{-13} A of NO^+ ions flowing to the first measuring electrode.

The cross section for the excitation of the 2p state of the hydrogen atom, $\sigma(2p)$, for this process was calculated from the formula[5)]

$$\sigma(2p) = \frac{4\pi}{\omega} I_{L_\alpha}{}^0 \Big/ \beta \frac{I^+}{e} N \Delta l, \qquad (2)$$

where $\sigma(2p)$ is the effective cross section for the excitation of the 2p state of the hydrogen atom; β is the coefficient of transmission of the L_α radiation by the lithium fluoride window; ω is the solid angle selected by the collimator of the ionization chamber; N is the number of gas atoms in 1 cm^3 in the collision chamber; I^+ is the current of protons entering the collision chamber; Δl is the length of a beam from which the L_α radiation reached the ionization chamber; e is the electronic charge.

[5)]Under our experimental conditions, there was equilibrium between the processes of formation and radiation by excited fast particles and, therefore, the coefficient allowing for the lifetime of the excited particles in Eq. (2) could be neglected.[11]

The measured effective cross section for the excitation of the 2p state of the hydrogen atom was 8.9×10^{-18} cm^2 for the process $H^+ + Ne \rightarrow H(2p) + Ne^+$ ($E_{H^+} = 16$ keV). The error in the measurement of this cross section did not exceed ±20%. This value of the cross section was used to normalize the excitation function of the L_α line, obtained for the same process using the vacuum monochromator. Next, using the known transformation coefficients, we calculated the cross sections for the excitation of the 2p states of the hydrogen atom in the remaining gases. The cross sections for the excitation of the 2s states were calculated from the measured ratios σ(2s)/σ(2p).

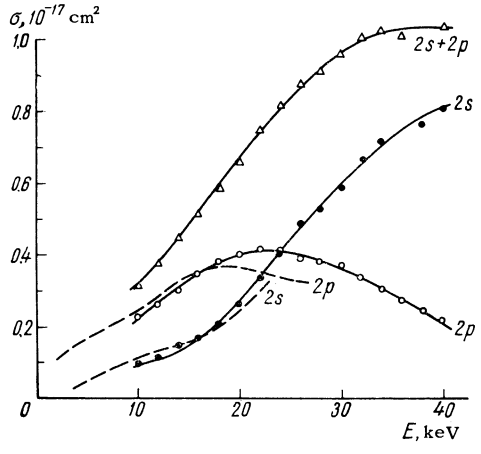

FIG. 4. Cross sections for the excitation of the 2s and 2p states and their sum for the process of the charge exchange of protons in helium, plotted as a function of the proton energy: ●– cross sections for the excitation of the 2s state; o – cross sections for the excitation of the 2p state; △ – sum of the excitation cross sections for the 2s and 2p states; dashed curves give the excitation cross sections of the 2s and 2p states reported in [7,14].

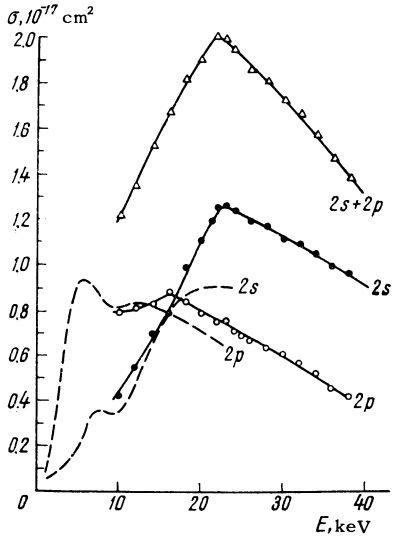

FIG. 5. Same as Fig. 4, but for protons in neon.

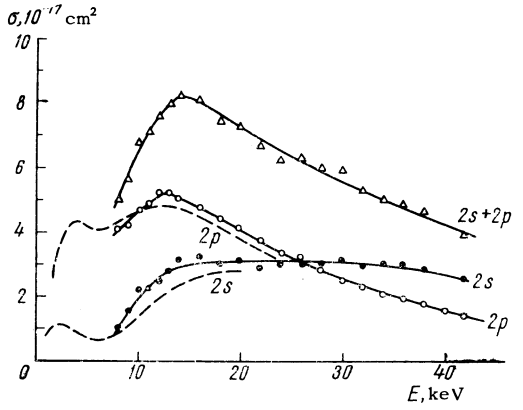

FIG. 6. Same as Fig. 4, but for protons in argon.

IV. RESULTS OF MEASUREMENTS

Figures 4–8 show the results of the measurements of the absolute values of the excitation cross sections of the 2s and 2p states of hydrogen atoms formed by the charge exchange of protons in helium, neon, argon, krypton, and xenon. The points on the curves represent the average values of several measurements. The reproducibility of the excitation functions of the L_α line lies within the limits of 5%. The same figures give the curves for the sums of the excitation cross sections of the 2s and 2p states of the hydrogen atom, and the curves for the cross sections σ(2s) and σ(2p) reported in [7, 14].

It follows from Figs. 4–8 that the excitation functions of the 2s and the 2p states have maxima in the investigated range of energies (the only exception is the charge exchange of protons in helium). The maximum values of the excitation cross sections of the 2s and 2p states [$\sigma_m(2s)$ and $\sigma_m(2p)$] and the total cross section for the excitation of the n = 2 level of the hydrogen atom [$\sigma_m(2s + 2p)$] by the charge exchange of protons are listed in the table. The values given in brack-

Maximum values of the cross sections for the excitation of the 2s and 2p states and of the n = 2 level of the hydrogen atom (10^{-17} cm^2)

Target	$\sigma_m(2p)$	$\sigma_m(2s)$	$\sigma_m(2s+2p)$
He	0.4 (22)	0.8 (> 40)	1.0 (~40)
Ne	0.89 (16)	1.29 (23)	2.0 (22)
Ar	5.3 (13)	3.0 (22)	8.4 (15)
Kr	7.5 (12,5)	5.3 (20)	12.5 (14)
Xe	11.6 (11)	8.7 (17)	19.4 (12)

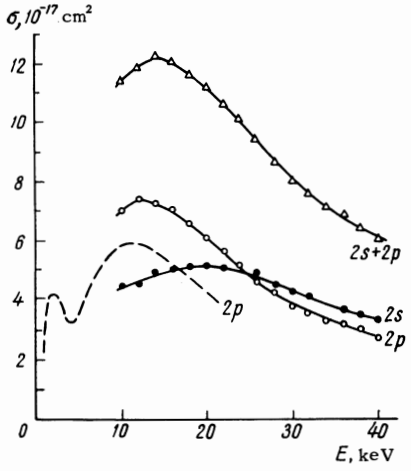

FIG. 7. Same as Fig. 4, but for protons in krypton.

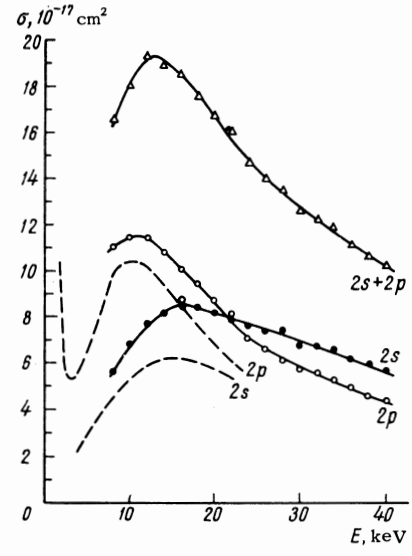

FIG. 8. Same as Fig. 4, but for protons in xenon.

ets in the table are the proton energies in keV at which the maximum cross sections are reached.

It is evident from Figs. 4–8 and the table that the maxima of the excitation cross sections of the 2s and 2p states of the hydrogen atom occur at different energies for the same gas. In the case of the charge exchange of protons involving the excitation of the 2s state, the maxima are reached at higher proton energy values compared with the formation of hydrogen atoms in the 2p state. The values of the proton energies at which the maximum values are reached for the excitation cross section of the 2p state depend on the nature of the gas: the higher the atomic number of the target atom, the lower this proton energy.

This dependence is less marked in the excitation of the 2s state. With the exception of helium, the values of the proton energies at which the excitation cross sections of the 2s state reach their maxima are similar for all the investigated gases. The maximum values of the excitation cross sections of the 2p state are larger than the corresponding cross sections of the 2s state in argon, krypton, and xenon. In helium and neon, the 2p state cross sections are smaller. Over the whole investigated range of proton energies, the absolute values of the excitation cross sections of the 2s and 2p states increase when the atomic number of the target particle is increased.

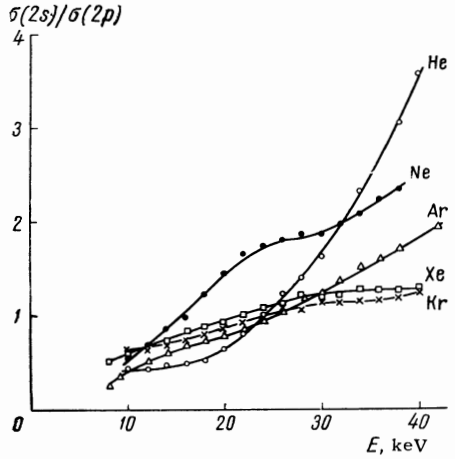

FIG. 9

Figure 9 shows the dependence of the ratio of the excitation cross sections of the 2s and 2p states of the hydrogen atom on the proton energy for all the investigated processes. As the proton energy is increased, the probability of the formation of hydrogen atoms in the 2s state increases for all the target gases. This is particularly clear in the case of the charge exchange of protons in helium. In the case of neon, the $\sigma(2s)/\sigma(2p)$ ratio has a plateau in the region of proton energies near 25 keV. At this energy, the velocity of the proton becomes equal to the velocity of the electron in the first Bohr orbit of the hydrogen atom. For the remaining gases, the ratio $\sigma(2s)/\sigma(2p)$ at this energy is close to unity, i.e., the capture of an electron by a proton in the charge exchange is equally likely for the 2s and 2p sublevels. At proton energies less than 25 keV, the probability of the excitation of the 2p sublevel is higher than the probability for the 2s sublevel, but on going over to heavier gases, the relative population of the 2s sublevel becomes somewhat greater. Conversely, at proton energies higher than 25 keV, the 2s sublevel of the hydrogen atom is predominantly excited in all the investigated gases. At these energies the relative population of the 2s sublevel increases on going over to lighter gases. The nature

of the dependence of the excitation cross sections on the azimuthal quantum number l for proton energies higher than 25 keV is in agreement with the conclusions reported in [15].

Jaecks, van Zyl, and Geballe[7] measured the excitation cross sections of the 2s state in the charge exchange of protons in inert gases (except krypton) in the proton energy range 1.5–23 keV. de Heer, van Eck, and Kistemaker[16] obtained the excitation cross sections of the 2p state for the charge exchange of protons in helium and neon (E_{H^+} = 5–35 keV). The results of our investigation agree best with the data reported in [7, 14]. The differences between the absolute values of the excitation cross sections $\sigma(2s)$ and $\sigma(2p)$ are within the limits of random error. This cannot be said about the results of de Heer et al.,[16] whose cross sections differed from ours by a factor of almost 3. However, the dependence of their cross sections on the proton energy is similar to that obtained in our study.

The cross sections for the excitation of the 2s and 2p states in the charge exchange of protons in helium have been calculated theoretically.[17] The cross sections $\sigma(2s)$ and $\sigma(2p)$, calculated in the Born approximation, decrease monotonically as the proton energy is increased in the range 10–40 keV, and the absolute values are in order of magnitude greater than the experimental ones. In the case of the charge exchange to the ground state, the position of the maximum, according to the Massey criterion, is given by the relationship

$$E_m = 3ma^2(\Delta E)^2, \qquad (3)$$

where E_m is the energy (in eV) corresponding to the maximum, a is some characteristic length (in Å) representing the range of interaction between the colliding particles, ΔE is the change in the internal energy of the particles taking part in the charge exchange (in eV), and m is the mass of the incident particle (in absolute mass units).

In the one-electron charge exchange, the length a has the same value of 7–8 Å for different interacting pairs (ion + atom). We calculated the value of a, using Eq. (3), from the positions of the maxima in Figs. 4–8 for the process of the charge exchange of protons in inert gases involving the formation of excited hydrogen atoms in the 2s and 2p states. We found that the values of a not only differed from gas to gas, but they were also different for the 2s and 2p states excited by the charge exchange in one and the same gas, although in practice $\Delta E(2s) = \Delta E(2p)$. In all the investigated cases, $a_{2S} > a_{2p}$. Both these quantities were larger for heavier gases. The values of a_{2S} ranged from 5.4 Å (for helium) to 8.5 Å (for xenon), and the values of a_{2p} ranged from 4 Å (for helium) to 6.9 Å (for xenon).

Thus, the Massey criterion cannot explain the positions of the maxima in the curves of the $\sigma(2s)$ and $\sigma(2p)$ excitation cross sections. Our cross sections were calculated on the assumption that the emitted L_α radiation was isotropic. Moreover, the influence, on the population of the 2s and 2p sublevels, of the cascade transitions from higher levels excited in the charge exchange was ignored. It can be shown qualitatively[7] that the possible departure from the isotropy of the L_α radiation and the cascade transitions cannot alter greatly the absolute values of the measured cross sections.[6)]

The authors are grateful to Professor V. M. Dukel'skiĭ for his constant help in this investigation. The authors are indebted to Yu. F. Bydin, who took part in the adjustment of the vacuum monochromator and in the first measurements with this instrument, and to V. B. Matveev who improved the stability of the ion beam.

[6)] Earlier,[18] we measured the cross sections for the excitation of the levels with n = 3–7 of the hydrogen atom by the charge exchange of protons in neon and helium. We estimated the contribution of the cascade transitions to the excitation cross section of the n = 2 level of the hydrogen atom. In the most unfavorable case (charge exchange of protons in helium, E_{H^+} = 30 keV), the contribution of the cascade transitions from the levels n = 3–7 was not more than 12%.

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