Ionization and excitation of Rydberg states in collisions of fast metastable helium atoms with He, Ne, and N_2

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The cross sections for ionization and excitation into Rydberg states ($n = 21$–27) were determined for fast (accelerated to 3.9 keV) metastable helium atoms colliding with He, Ne, and N_2. An analysis of the collisionally excited helium atoms in terms of their principal quantum number was carried out using for the ionization by an electric field a system capable of separation in accordance with n and was calibrated using signals from Rydberg states excited selectively by laser radiation.

INTRODUCTION

The experimental values of the cross sections of elementary collision processes involving neutral atoms and covering the energy range 0.1–1000 keV are of interest because they can be used to model some phenomena which occur under laboratory conditions and in outer space,[1] and also to check the precision of various models and methods used in the theory of atomic collisions. The latter task is particularly important at moderate collision energies (usually below 100 keV) where the Born approximation for the cross sections of collisional processes gives results greatly at variance with the experimental data and the precision of the more complex theoretical methods being developed at present requires a careful experimental check.[1,2]

We shall report an investigation of two elementary collisional processes involving metastable helium atoms (He*) accelerated to 3.9 keV:

$$\mathrm{He}^*+\mathrm{X}\rightarrow\mathrm{He}^+ + \mathrm{X}(\Sigma)+e^-, \quad (1)$$

$$\mathrm{He}^*+\mathrm{X}\rightarrow\mathrm{He}(nR)+\mathrm{X}(\Sigma), \quad (2)$$

where X is an atom or a molecule (He, Ne or N_2) in its unexcited (ground) state, whereas $\mathrm{X}(\Sigma)$ is the same atom or molecule in any excited, unexcited, or ionized state, including the state of dissociation of the N_2 molecule; $\mathrm{He}(nR)$ is helium in a Rydberg state with the principal quantum number n.

The present authors are not aware of any experimental data on the cross section of the process (1) in the energy range < 25 keV. Estimates of this process given in Ref. 3 for the range of energies of interest to us represent a theoretical extrapolation of the experimental results obtained at higher energies.

There are also practically no data on the excitation cross sections of Rydberg states by the process (2) and this applies to collisions of any neutral atoms at any collision energies. This is obviously due to the very small values of these cross sections, so that it is very difficult to distinguish the useful signal from the background and record it. We can mention here the investigations reported in Refs. 4 and 5, where the luminescence signal was used to find the excitation cross sections of various states reached as a result of collisions of helium atoms. The authors of Refs. 4 and 5 were able to determine the excitation cross sections of the n^3D states for $n \leqslant 16$ when atoms were in the ground electronic state and for $n \leqslant 12$ when one of the atoms was initially in a triplet metastable state. The fullest theoretical investigation of the excitation of Rydberg states in the specific case of collisions of two hydrogen atoms was reported in Ref. 1.

It therefore follows that it would be interesting to determine the cross sections of these processes because other experimental data are absent or unsatisfactory.

Our experiments were carried out using apparatus intended for laser photoionization detection of rare isotopes in

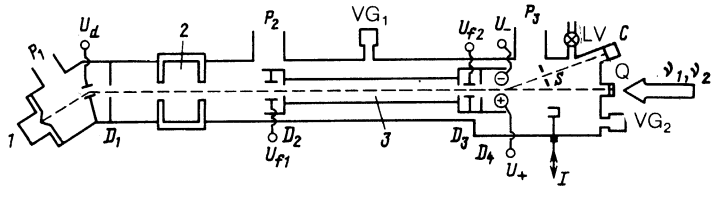

FIG. 1. Schematic diagram of the apparatus: 1) source of helium ions; 2) charge-exchange chamber; 3) field-free region; D_1–D_4 are the stops; U_{f1} and U_{f2} are the voltages across the filtering capacitors f_1 and f_2; U_d is the voltage across the deflector d; U_+ and U_- are the voltages across the ionizing cylinders; S is the variable-width slit; C is the secondary-electron multiplier; VG_1 and VG_2 are the pressure gauges; I is the meter for measuring the current; Q is the quartz substrate; LV is the leak valve; P_1–P_3 are the vacuum pumps.

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an accelerated atomic beam. This isotope detection method[6] is based on isotopically selective laser excitation of atoms of a rare isotope to a Rydberg state, followed by subsequent ionization in an electric field; the starting point is a fast atomic beam formed by neutralization of a beam of ions accelerated by the application of a potential U_a. In the case of collinear excitation the absorption spectrum of such an atomic beam exhibits an additional kinematic isotopic shift which is due to the difference between the velocities of motion of the isotopes which have the same kinetic energy $E_k = eU_a$. This additional isotopic shift is much larger than the natural shift so that it is possible to ensure an extremely selective multistage laser excitation of atoms of any isotope to a Rydberg state. Such Rydberg atoms are then ionized by an electric field and the ions then formed are detected.

The example of the rare iostope ^{3}He was used in Ref. 7 to show that the detection selectivity is limited by the background signal due to two collisional processes involving atoms in an accelerated beam and molecules of the residual gas. These processes are the collisional ionization of the accelerated atoms and the collisional excitation of these atoms to Rydberg states. We shall report a determination of the cross sections of these processes from the dependences of the background signal on the pressure of the gases admitted into the system.

APPARATUS

We used apparatus intended for laser photoionization detection of the rare isotope ^{3}He, which was described in detail in Ref. 7 and shown schematically in Fig. 1. A beam of accelerated (to 3.9 keV) helium atoms was created by a gas-discharge ion source with a hot cathode 1 and was collimated by a single electrostatic lens; it was deflected by a deflector d in order to remove the neutral component. The deflected ion beam was directed to a charge-exchange chamber 2 containing potassium vapor. The temperature of this chamber was $T = 155$ °C and then the coefficient representing charge exchange resulting in conversion of the accelerated ions into atoms amounted to $k = 0.40 \pm 0.05$. About three-quarters of the atoms generated in this way were in a triplet metastable state.[8] The accelerated atomic beam was filtered in a field generated by a capacitor f_1 in order to remove the ions which did not undergo the charge exchange and also those highly excited atoms which could be created by the process of charge exchange. The electric field in this capacitor $E_{f1} = 5$ kV/cm was sufficient for the ionization of Rydberg states with principal quantum numbers $n \gtrsim 17$ ($E_i \sim E_{cr} = 1/16n^4$).[9] This filtering capacitor removed ions completely on the axis of the apparatus. (For example, when the charge exchange chamber was cold and the voltage U_{f1} was applied, no ions reached a detector C.)

After this filtering capacitor the accelerated atomic beam reached a field-free region 3. The laser radiation applied in this region was used to excite the triplet metastable He atoms from the 2^3S state to various n^3S and n^3D states via an intermediate 3^3P level; this was done by employing two pulsed dye lasers. The laser radiation was directed opposite to the accelerated atomic beam. At the exit from the field-free region (closed by a stop D_3 with an aperture 2 mm in diameter) the atomic beam reached a region where an electric field was established by an ionizer consisting of two cylinders and where the ionization of the Rydberg atoms took place. The same field deflected the resultant ions to a variable-width slit S behind which a secondary-electron multiplier was located. The ions and the highly excited atoms formed in the field-free region could be extracted from the beam before they reached the ionizer and this was done by a second filtering capacitor f_2.

The signal from the secondary-electron multiplier was analyzed by a gated pulse-counting system or, at high signal amplitudes (exceeding 30 pulses/μs), by a recording system which integrated the current passing through the multiplier after each laser pulse. The signal was then averaged by a gated voltmeter and delivered to an XY plotter.

The ion beam current I was measured with a Faraday cylinder and the neutral particle flux was determined utilizing secondary electron emission. The second electron emission coefficient of the tantalum bottom of a sensor based on secondary electron emission was $\eta_i = 0.43 \pm 0.04$. According to the published data,[10] when the energy of the incident particles was 3.9 keV, the secondary electron emission coefficient for the He atoms in the ground electronic state should be 1.07 ± 0.2 times greater than for ions, i.e., it should be 0.46 ± 0.1. We assumed that the secondary electron emission coefficient for the atoms in the metastable states η_m agreed with the value just quoted (within the limits of its error). The flux of metastable atoms crossing a stop D_3 amounted to $I \approx 5 \times 10^{10}$ particles/s.

The efficiency of the system for recording the ions was determined by deflecting the ion current (10^{-12} A), measured using the Faraday cylinder, to the secondary electron multiplier. The efficiency of the recording process, defined as the ratio of the number of counted ions to the total number of ions directed to the multiplier, amounted to 0.40 ± 0.02.

The pressures were measured using gas-discharge VG_1 (PMI-2) and magnetic-discharge VG_2 pressure gauges. Correction coefficients[11] were applied when dealing with the pressures of different gases. The error in the absolute measurements of the pressure in the case of our gauges amounted to $\pm 35\%$ for VG_1 and to $\{^{+80}_{-50}\%$ for VG_2.

A leak valve LV was used to admit various gases into the vacuum system. The vacuum pump P_2 was then shut off and we assumed that the pressure of the admitted gas throughout the volume between the stop D_1 and the filtering capacitor f_2 did not vary by more than a factor of 1.5 and could be determined using the vacuum gauge VG_1 with an absolute error of $\pm 50\%$.

The pressure measurements in the region of ionization of Rydberg atoms were carried out using the gauge VG_2. Since the pump P_3 and the leak valve LV were not shut off, the error due to inhomogeneity of the distribution of the pressure in these measurements could reach 100%.

IONIZATION OF ATOMS IN RYDBERG STATES

The system for the ionization of Rydberg atoms in an electric field of two cylinders, used in our experiments, was capable of separation of atoms in accordance with the principal quantum number n. In fact, different Rydberg states were ionized by different electric fields, i.e., at different points of their path, so that the resultant ions were deflected through different angles.

We plotted in Fig. 2 the dependences of the signal on the

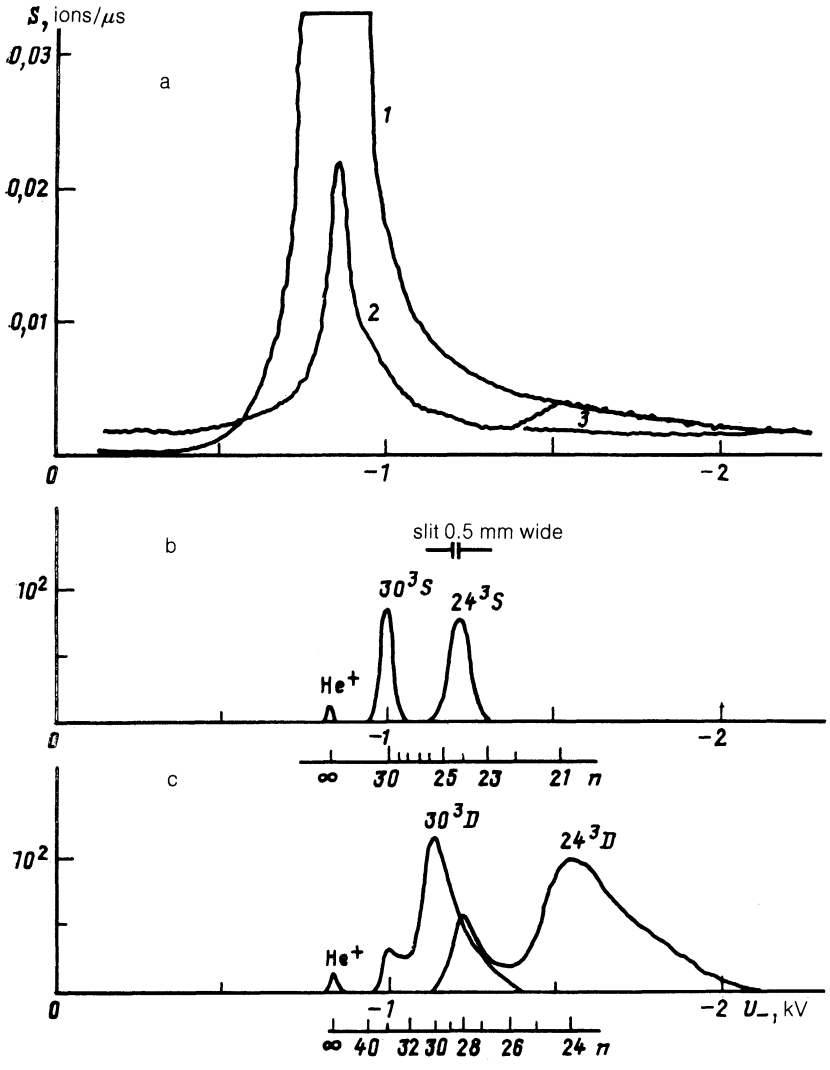

FIG. 2. a) Dependence of the background signal on U_-, obtained for different voltages across the second filtering capacitor U_{f2} ($U_+ = 750$ V): 1) 0 kV; 2) -1 kV; 3) -2 kV. b), c) Dependences of the ion signal obtained on laser excitation of the states 30^3S, 24^3S (b) and 30^3D, 24^3D (c) on U_- ($U_+ = 750$ V, $U_{f2} = 0$). The positions of the maxima of the signal obtained for various n^3S and n^3D states are shown below the abscissas.

voltage U_- applied to one of the ionizing cylinders in the case of excitation of the 24^3S and 30^3S states (Fig. 2b) and the 24^3D and 30^3D states (Fig. 2c). A peak observed in all the dependences at $U_- = -850$ V was due to the appearance of ions in the field-free region as a result of excitation of metastable atoms by laser radiation in the first stage; they were excited by the continuum of states via an intermediate 3^3P level and this was followed by collisional ionization of the accelerated helium atoms in the field-free region. The ionization of the atoms excited to different n^3S states gave rise to identical peaks representing adiabatic evolution[9] of an atom characterized by $|m_l| = 0$ in an electric field. In the case of atoms excited to n^3D states we observed two peaks, the first of which was associated with adiabatic evolution in an electric field of the atoms characterized by $|m_l| = 0$ and 1, which coincided with the peak for n^3S states; the second peak was mainly due to adiabatic evolution in an electric field of atoms with $|m_l| = 2$ (for details see Ref. 7). Figure 3 shows the dependences on the principal quantum number n of the voltages at which a maximum of the signal for n^3S states was observed (curve 1) and where a maximum of the signal for the second peak of the n^3D states was recorded (curve 2).

The top part of Fig. 4 gives the dependences of the photoion signal for the 32^3D ($U_- = -970$ V) and 26^3D ($U_- = -1120$ V) states on the filtering voltage U_{f2}. The lower parts of the same figure give the dependences on U_{f2} of the collisional background obtained under the same conditions, but in the absence of laser radiation ($U_- = -970$ V for Fig. 4a and $U_- = -1120$ V for Fig. 4b). Clearly, at voltages U_{f2} amounting to 0.5 kV (Fig. 4a) and 1.1 kV (Fig. 4b) there was a strong reduction in the photoion signal to zero and of the background by a factor of about 2. A further increase in U_{f2} did not alter the background level. Such a reduction in the signal and background was due to the ionization of Rydberg atoms in the field of the filtering capacitor f_2. The signal represented the Rydberg atoms excited by the incident laser radiation, whereas the background represented the atoms created by collisions with molecules of the residual gas in the field-free region.

We plotted in Fig. 2a the dependences of the background on the voltage U_- when U_{f2} amounted to 0, -1, or -2 kV. Clearly, in the range -1800 V $< U_- < -950$ V approximately half of the background signal was associated with the excitation of the Rydberg states in the course of the collisions and it disappeared on application of the voltage U_{f2}. The remaining background was due to the collisional ionization of the metastable atoms in the electric field of the ionizing cylinders in the same place where the ionization of the laser-radiation-excited "signal" Rydberg atoms took

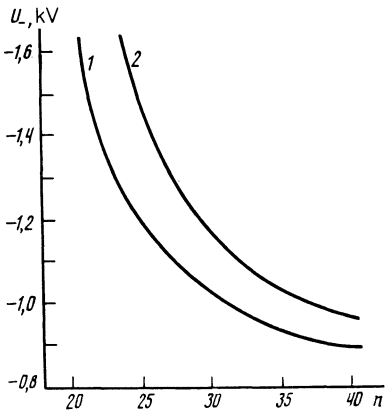

FIG. 3. Experimental dependences of the voltage U_-, corresponding to a maximum of the signal of the n^3S (curve 1) and n^3D (curve 2) states, on the principal quantum number n.

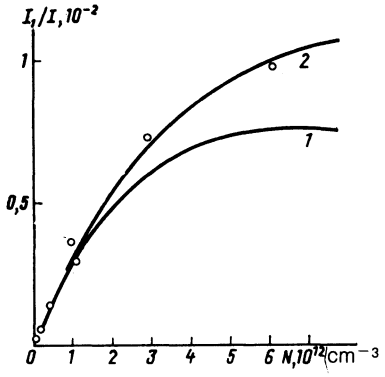

FIG. 5. Dependences of the fraction of ions I_1/I in the atomic beam on the density N of helium atoms in the system; the points are the experimental values; 1) calculated dependence obtained by substituting $\sigma_{10} = 7.2\times 10^{-16}$ cm^2 (Ref. 12) and $\sigma_{0^*0} = 5.0\times 10^{-16}$ cm^2 (Ref. 13); 2) calculated for values for σ_{10} and σ_{0^*0}, reduced by 30% compared with those just given.

place. In this case the signal ions and those formed by collisions were indistinguishable and were recorded in the same way by the counting system.

An increase in the electric field intensity in the filtering capacitor f_1 from 3 to 9 kV/cm, corresponding to the ionization of the Rydberg states with the principal quantum numbers n from about 18 to 14, produced no changes in the background signal. Hence, the Rydberg atoms with low values of n, which were not filtered off by the capacitor f_1, made in fact no contribution to the observed background signal. Moreover, the excited states with low values of n exhibited faster radiative decay.[9]

DETERMINATION OF THE IONIZATION CROSS SECTION OF FAST METASTABLE HELIUM ATOMS COLLIDING WITH VARIOUS GASES

We determined the ionization cross section of metastable helium atoms (i.e., the electron-loss cross sections) by selecting the voltages across the ionizing cylinders ($U_+ = 750$ V, $U_- = -850$ V) in such a way that all the ions formed in the field-free region were deflected to the secondary-electron multiplier. The circles in Fig. 5 represent the experimental dependences of the fraction of the ions in the beam at the exit from the field-free region I_1/I on the density N of the helium atoms in the system. At low values of N the dependence was linear and it obeyed the simple relationship

$$I_1/I = \sigma_{0^*1} N l,$$

where $l = 110$ cm is the length of the field-free region and σ_{0^*1} is the collisional ionization cross section of the metastable helium atoms. Table I gives the values of σ_{0^*1} obtained for various gases from the linear parts of the experimental dependences. The absolute error of these measurements was $\{^{+100}_{-50}\}$%. The relative error in determination of the cross section σ_{0^*1} amounted to $\pm$ 20% for the various gases.

It is clear from Fig. 5 that at high values of N the experimental dependence ceased to be linear. This could be explained by charge exchange experienced by the resultant ions (σ_{10}) and by deexcitation of the metastable helium atoms (σ_{0^*0}) colliding with the target atoms. The collisional ionization cross section of helium atoms in the ground state σ_{01} was approximately an order of magnitude less than σ_{0^*1} (Ref. 12) and in the range $N < 6\times 10^{12}$ cm^{-3} when the fraction of the atoms in the ground state in the atomic beam was less than half, we could ignore this process.

In this approximation the fraction of the metastable atoms in the accelerated beam entering the field-free region is

$$\frac{I_{0^*}}{I}(x=0) = \frac{k\exp[-(\sigma_{10}l_1+\sigma_{0^*0}l_2)N]}{1-(1-k)\exp[-\sigma_{10}(l_1+l_2)N]},$$

where I_{0^*} $(x = 0)$ is the flux of metastable atoms at the entry to the field-free region; $k = 0.4$ is the coefficient representing

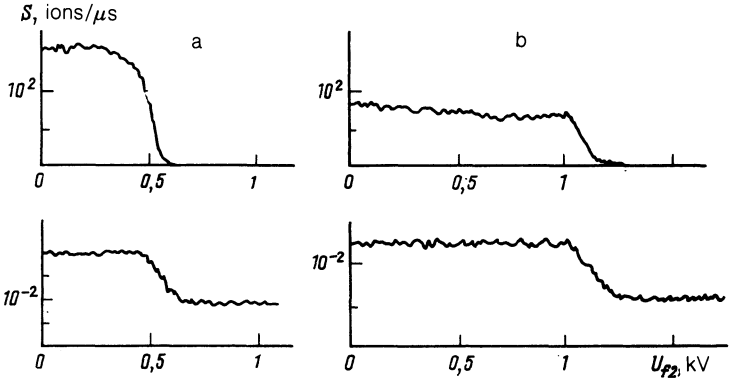

FIG. 4. Dependences of the ion signal obtained on laser excitation of the 32^3D (a) and 26^3D (b) states (upper part of the figure) and of the background signal (lower part of the figure) on the voltage U_{f2} applied to the second filtering capacitor in two cases $U_- = -970$ V (a) and -1120 V (b).

TABLE I. Experimental cross sections of the collisional ionization (σ_{0^*1}) and of the collisional excitation to $n = 24$ Rydberg states (σ_{0^*24}) of metastable (75% triplet) helium ions of 3.9 keV energy colliding with He, Ne and N_2.

Target atom (molecule)	σ_{0^*1}, 10^{-17} cm^2	σ_{0^*24}, 10^{-20} cm^2
He	3,3	8,3
Ne	1,5	3,8
N_2	8,8	16

charge exchange of the ions in the charge-exchange chamber; $l_1 = 30$ cm and $l_2 = 40$ cm are the distances from this chamber to the stops D_1 and D_2, respectively. In the field-free region this quantity decreases in accordance with the law

$$\frac{I_{0^*}}{I}(x)=\frac{I_{0^*}}{I}(x=0)\exp[-(\sigma_{0^*0}+\sigma_{0^*1})Nx],$$

where x is the distance from the entry to the field-free region.

The fraction of the ions in the beam I_1/I in the field-free region can be found from the equation

$$d\frac{I_1}{I}(x)=\sigma_{0^*1}N\frac{I_1}{I}(x)dx-\sigma_{10}N\frac{I_1}{I}(x)dx.$$

The relative fraction of the ions reaching the detector is then

$$\frac{I_1}{I}=\exp(-\sigma_{10}Nl_3)\,\frac{I_1}{I}(x=l),$$

where the factor $\exp(-\sigma_{10}Nl_3)$ is due to charge exchange experienced by some of the ions in the field of the ionizing cylinders ($l_3 \approx 5$ cm) before they are deflected to the detector. We finally have

$$\frac{I_1}{I}=\frac{k\sigma_{0^*1}}{\sigma_{10}-\sigma_{0^*0}-\sigma_{0^*1}}\,\frac{\exp[(\sigma_{10}-\sigma_{0^*0}-\sigma_{0^*1})Nl]-1}{1-(1-k)\exp[-\sigma_{10}(l_1+l_2)N]}\times\exp\{-[\sigma_{10}(l_1+l+l_3)+\sigma_{0^*0}l_2]N\}.$$

Curve 1 in Fig. 5 gives the dependence of I_1/I on the density N of the helium atoms obtained by substituting the values $\sigma_{10} = 7.2\times10^{-16}$ cm^2 (Ref. 12) and $\sigma_{0^*0} = 5.0\times10^{-16}$ cm^2 (Ref. 13). The processes of deexcitation of the metastable atoms and of the charge exchange of the ions influence approximately equally the nature of saturation of the dependences of I_1/I on N. The degree of their influence can be estimated using a calculated curve 2 obtained by substituting the values of σ_{10} and σ_{0^*0} reduced by 30% compared with those given above and taken from Refs. 12 and 13.

As pointed out already, the ionization of the fast metastable helium atoms in the electric field of the ionizing cylinders was represented by a part of the background signal which did not disappear on application of U_{f2} (Fig. 2a). The pressure dependences of this background signal could be used also to determine the cross section σ_{0^*1}. However, this could be done only knowing the length of the region Δl subject to the electric field, which was crossed by ions before reaching the slit S and being recorded by the secondary electron multiplier. At low values of N the background signal was then described by the simple expression:

$$I_1/I=\sigma_{0^*1}N\Delta l.$$

Assuming that the angles of deflection of the ions by the electric were not too large (in our case the relevant value was $\alpha = 20.3°$), we readily obtained the following expression for Δl:

$$\Delta l=\frac{2U_a(1+\operatorname{tg}\alpha)}{E_i}\Delta\alpha,$$

where $\Delta\alpha$ is the angle subtended by the slit as observed from the point of ionization and E_i is the electric field in the region from which ions arrived. Using curve 1 in Fig. 3 we could assign the principal quantum number n of the Rydberg n^3S states of the ions which crossed the slit for a given values of U_- and this could be done for any voltage in the range $-1600\ \text{V} < U_- < -900\ \text{V}$. Since ionization of the n^3S states was an adiabatic process,[7] the intensity of the electric field in the region where the ions were formed could be calculated from the expression $E_i \sim E_{cr} = 1/16n^4$. Substituting the value obtained in this way in the expression for Δl, we could find the length Δl for any value of U_-.

The cross sections σ_{0^*1} obtained in this way from the dependences of the background signal on the pressure of He and N_2 in the system are given in Table II for different values of U_-. These cross sections were determined much less accurately than the cross sections σ_{0^*1} deduced from the ion

TABLE II. Collisional ionization cross sections (σ_{0^*1}) of accelerated He atoms colliding with He and N_2, deduced from the pressure dependences of the collisional background originating from the region of application of the electric field using different values of U_-.

	U_-, V					
	−1550	−1360	−1240	−1140	−1070	−1000
$\sigma_{0^*1}^{N_2}$, 10^{-17}, cm^2	9,0	9,0	9,2	10,0	9,4	10,6
$\sigma_{0^*1}^{He}$, 10^{-17} cm^2	2,7	3,0	3,1	3,4	3,5	3,6

signal in the field-free region (Table I) which was primarily due to the larger error in the determination of the pressure in the ionization region. However, the good agreement between the results obtained demonstrated additionally that we interpreted correctly the background which was retained on application of U_{f2} (Fig. 2a) as the result of collisional ionization of the accelerated atoms. In the range $U_- > -1070$ V the values of $\sigma_{0^*_1}$ were somewhat overestimated because the electric field of the filtering capacitor f_2 penetrated into the ionization region through the stop D_4.

We are not aware of any published experimental values of $\sigma_{0^*_1}$ for collisions characterized by energies of several kiloelectron-volts. However, our results do agree with those theoretical estimates of $\sigma_{0^*_1}$ for helium which are given in Refs. 3 and 14. The ratios of the cross sections $\sigma_{0^*_1}$ obtained by us for different gases are in agreement with those determined for collision energies 50 keV and reported in Ref. 15.

Although the fraction of the metastable atoms in the 2^1S state in an accelerated beam can be small (less than one-quarter in Ref. 8), the ionization cross section can be considerably greater than for the atoms in the 2^3S states.[14] Then, the presence of the singlet metastable component of the beam can have a significant influence on the value of $\sigma_{0^*_1}$. The published literature includes only the data on the collisional deexcitation of metastable states. The cross section of this process is 1.6 times greater for singlet than for triplet atoms.[13]

DETERMINATION OF THE CROSS SECTIONS OF COLLISIONAL EXCITATION OF FAST METASTABLE He ATOMS TO RYDBERG STATES

As shown already, a part of the signal disappearing on application of the voltage U_{f2} (see Figs. 2a and 4) was due to the excitation of the metastable helium atoms to the Rydberg states by collisions with molecules of the residual gas in the field-free region. The cross section of this process was determined by recording the dependences of the "Rydberg" signal on the gas pressure in the target for several values of U_-. The signal was found to be proportional to the width of the slit S. The excitation cross sections of the Rydberg states could be determined for collisions with different gases provided we knew which states and to what degree contributed to the signal obtained using a given slit width. It was impossible to identify experimentally the Rydberg states excited in the collisions and find how these states were ionized in the electric field. Therefore, we shall consider two extreme cases of adiabatic and diabatic ionization of collisionally excited atoms.

Let us assume that atoms excited in collisions are ionized mainly by the adiabatic process. In this case even when various Stark sublevels of the hydrogen-like series are excited, atoms are ionized in fields of intensities close to $E_{cr} = 1/16n^4$, i.e., in fields similar to those causing ionization of the signal n^3S states. Next, the dependence 1 in Fig. 3 can be used to determine the principal quantum number of those excited states which make the main contribution to the signal measured at a given value of U_-. The fraction of the ions which appear as a result of ionization of the background states with a given value of n and which cross the slit can be found from the expression

$$dn = \frac{\partial n}{\partial S}\,dS = \frac{\partial n}{\partial U_-}\frac{\partial U_-}{\partial S}\,dS,$$

where n is the principal quantum number, U_- is the voltage across the ionizing cylinder, and S is the slit width. The quantity $\partial U_-/\partial S$ was deduced from the broadening of the signal peaks when the slit was widened. In the working range of U_1 it amounted on the average to 32.5 V/mm. The quantity $\partial U_-/\partial S$ for the adiabatic ionization case was determined from curve 1 in Fig. 3. Having found the value of dn in this way, we were able to deduce the signal associated with the ionization of the states with a given value of n using the expression $I_n = I_{ph}/dn$, where I_{ph} is the experimentally determined signal. The circles in Fig. 6 represent the dependence of I_{21S} ($U_- = -1600$ V) on the density N of the helium atoms in the field-free region. Clearly, at low densities the dependence was linear and then reached saturation. Similar dependences could be plotted also using curve 2 in Fig. 3 on the assumption of predominantly diabatic evolution of the collisionally excited atoms in an electric field. The cross sections for the collisional excitation of the states with the principal quantum number n were found from the linear parts of these dependences using the expression $\sigma_{0^*_n} = I_n/INl$.

Accurate measurements of the cross sections $\sigma_{0^*_n}$ could be made only for values of U_- ranging from -1070 to -1600 V, corresponding to the recorded range of n from 21 to 27 in the adiabatic case and/or from 24 to 32 in the case of diabatic evolution of the Rydberg atoms excited by collisions. In the range $U_- < -1600$ V the ions formed by the ionization of the Rydberg states were deflected again by the electric field of the ionizing cylinders and failed to reach the detector. At voltages $U_- > 1070$ V it was impossible to separate correctly the components of the signal (collisionally excited Rydberg atoms and collisional ionization) because the ions reaching the detector were formed at the entry to the electric field of the ionizing cylinders, which was influenced by the electric field of the filtering capacitor f_2 penetrating through the stop D_4.

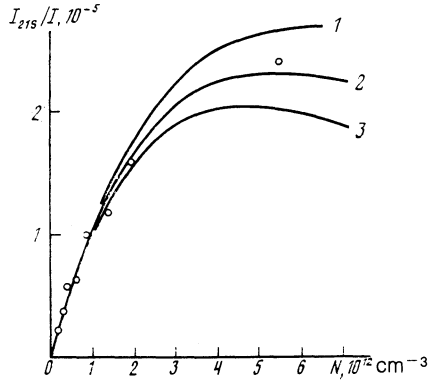

FIG. 6. Dependences of the ratio of the background ions formed as a result of ionization of the states with $n = 21$ on the assumption of adiabatic evolution, to the total number of particles in atomic beam on the density N of the helium atoms in the system; the circles are the experimental values. The curves are calculated substituting $\sigma_{0^*_0} = 5.0\times10^{-16}$ cm^2 (Ref. 13) and $\sigma_{10} = 7.2\times10^{-16}$ cm^2 (Ref. 12), and also our values of $\sigma_{0^*_1} = 3.3\times10^{-17}$ cm^2, $\sigma_{0^*_{21}} = 10.8\times10^{-20}$ cm^2, and $\sigma_d = 1\times10^{-15}$ cm^2 (curve 1), $\sigma_d = 1.5\times10^{-15}$ cm^2 (curve 2), and $\sigma_d = 2\times10^{-15}$ cm^2 (curve 3).

The values of the collisional excitation cross sections σ_{0^*n} of the Rydberg states with the principal quantum numbers $n = 24$–27 were found to be practically the same for the adiabatic and diabatic mechanisms of the ionization of the collisionally excited atoms. We listed in Table I the values of the cross sections σ_{0^*24}. The absolute error in these measurements was $\{^{+200}_{-65}\%$. The relative error for the various gases was $\pm 30\%$.

An increase in n reduced rapidly the cross sections σ_{0^*n}. Assuming that the reduction obeyed the power law $\sigma_{0^*n} \propto n^{-\alpha}$, we found that the power exponent was $\alpha = 3 \pm 1.2$. A theoretical analysis gave $\sigma_{0^*n} \propto n^{-3}$ (Refs. 16 and 17).

The deviation of the experimental dependences from linearity (Fig. 6) observed at high values of N was associated not only with the processes discussed in the preceding section (σ_{10} and α_{0^*0}), but also with collisional deexcitation of the Rydberg atoms (σ_d). The fraction of the Rydberg atoms in the accelerated beam I_n/I could be found in the field-free region by solving the equation

$$d\frac{I_n}{I}(x) = \sigma_{0^*n} N \frac{I_{0^*}}{I}(x)\,dx - \sigma_d N \frac{I_n}{I}(x)\,dx,$$

where x is the distance from the entry to the field-free region and I_{0^*}/I is the fraction of the metastable atoms in the beam. Then, the relative fraction of the Rydberg atoms with a given value of n could be found at the exit from the field-free region using the expression

$$\frac{I_n}{I} = \frac{k\sigma_{0^*n}}{\sigma_d - \sigma_{0^*0} - \sigma_{0^*1}} \frac{\exp[(\sigma_d - \sigma_{0^*0} - \sigma_{0^*1})Nl] - 1}{1 - (1-k)\exp[-\sigma_{10}(l_1+l_2)N]} \times \exp[-(\sigma_{10}l_1 + \sigma_{0^*0}l_2 + \sigma_d l)N].$$

Figure 6 shows the calculated dependences of I_{21S}/I on the density N of the helium atoms plotted for different values of σ_d. We used $\sigma_{0^*0} = 5.0 \times 10^{-16}$ cm^2 (Ref. 13) and $\sigma_{10} = 7.2 \times 10^{-16}$ cm^2 (Ref. 12), as well as the values of $\sigma_{0^*1} = 3.3 \times 10^{-17}$ cm^2 and $\sigma_{0^*21} = 10.8 \times 10^{-20}$ cm^2 obtained in the present study. Clearly, the change in σ_d had little effect on the nature of saturation, but it prevented us from determining σ_d from the nature of such saturation. We could simply say that σ_d exceeded the value of σ_{10} and/or σ_{0^*0} by a factor not exceeding 2–3. A similar estimate of σ_d was obtained from an analysis of the reduction in the photoionic signal on increase in the helium pressure in the system when the Rydberg states were excited by laser radiation.

As pointed out already, excitation of the Rydberg states as a result of collisions of neutral atoms has been investigated experimentally only using luminescence signals.[4,5] These luminescence experiments did not allow us to determine the total cross section for the excitation to all the quantum states with a given value of n. Instead, measurements were made of the cross sections of the excitation of the states with a definite value of the orbital momentum l.

Theoretical relationships between the excitation cross sections of the Rydberg states and the ionization cross section were obtained in Ref. 17. However, these relationships could be checked only if the experimental energy dependence of one of the cross sections was available and, moreover, if we knew several parameters for the He* + He system, which were not available to us.

CONCLUSIONS

We determined the cross sections for the collisional ionization and excitation of accelerated metastable (75% triplet) helium atoms to Rydberg states. We demonstrated that one could use the method of selective ionization by an electric field to separate the signals representing the various Rydberg states formed as a result of collisions of neutral atoms. This approach could also be used to find the cross sections for the collisional excitation of Rydberg states by collisions of any neutral atoms and this could be done in a wide range of energies.

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