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IONIZATION PRODUCED IN COLLISIONS BETWEEN ALKALI METAL ATOMS AND GAS MOLECULES

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The total cross sections σ_- for the production of free electrons, the ionization cross sections σ_+, and the stripping cross sections σ_l are measured for single collisions between Li, Na, K, and Cs atoms and the atoms of all inert gases and H_2 and N_2 molecules. In the investigated energy range 3—30 keV all these cross sections increase continuously with the velocity of the incident particles. As a rule σ_-, σ_+, and σ_l for a given alkali metal atom also increase with the atomic number of the target particles. This rule breaks down when the atomic numbers of the colliding particles are close (e.g., Na-Ne, K-Ar, and Cs-Xe), in which case the ionization cross sections are considerably larger. The stripping cross sections for Li atoms in He, H_2, and N_2 are compared with the available experimental data, and the $\sigma_-(v)$ curves are compared with Firsov's theoretical curve. It is shown that the theory yields excessive values of σ_- for ionization processes involving alkali metal atoms. Factors affecting the ratio between σ_+ and σ_l are discussed.

1. INTRODUCTION

IONIZING collisions between fast multi-electron atoms and gas atoms or molecules have been investigated mainly with fast inert-gas atoms.[1,2] Another important group of elements, the alkali metals, has received insufficient attention. Cross sections for the ionization of gas atoms and molecules by alkali metal atoms have not been measured; the existing experimental data on the ionization (stripping) cross sections of alkali metal atoms are confined to relatively low energies (150—2200 eV).[3,5] Stripping at high energies has been investigated only for lithium atoms in He and H_2 at 5—22 keV,[6] and in He, H_2, and N_2 at 10—475 keV.[7]

The present work is a comprehensive investigation of ionizing collisions between alkali metal atoms and gas atoms or molecules in the range 3—30 keV. The measured cross sections may be useful in connection with different applications of the physics of atomic collisions (corpuscular diagnosis of plasmas, ion motors, astrophysics, and mass-spectrometry). It will also be possible to test Firsov's statistical theory,[8] which has been the only attempt to calculate ionization cross sections for the collisions of multi-electron atoms in the keV region.

2. EXPERIMENT

The cross sections for the ionizing processes were measured by the condenser technique.[1,9] Beams of fast alkali metal atoms were obtained through the charge change of the alkali metal ions in nitrogen. To obtain K^+ and Cs^+ ions we used the surface ionization source described in [10]; Na^+ and Li^+ were obtained from a thermionic source. The latter was a tungsten coil wound around an alundum rod which was placed in a tantalum container. A suspension of a powder prepared from aluminosilicates of alkali metals was deposited on the coil and the rod; the preparation of this powder is described in [11]. The porous alundum surface ensured absorption of the suspension and a lengthy working period (~200 hours) of the source. We have described the other parts of our experimental apparatus in [1,9].

We know [1] that the condenser technique can be used to determine independently the total cross sections for the production of free electrons (σ_-) and positive ions (σ_+). For atom-atom collisions σ_+ is the cross section for pure gas ionization, and the stripping cross section is $\sigma_l = \sigma_- - \sigma_+$. Direct measurement of σ_l by registering fast ions, produced in a collision chamber, in the detector of a mass analyzer (Fig. 1 of [12]) is less

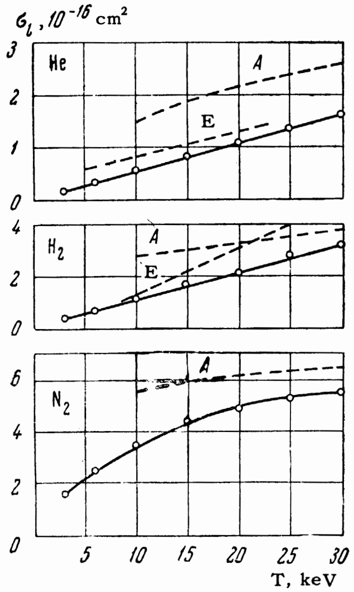

FIG. 1. Stripping cross sections of fast Li atoms in He, H_2, and N_2 were measured in the present work (solid curves) and in [6,7] (dashed curves): E for [6] and A for [7].

accurate because these ions are scattered appreciably in the gas. As in our other investigations, the fast neutral atom beam intensity was registered by means of secondary electron emission. The fast atom flux in the collision chamber, expressed in terms of the equivalent current, was $\sim 10^{-9}$ A, while the currents to the measuring plates of the condenser were two or three orders of magnitude smaller. The currents were measured with an ÉMU-3 electrometer amplifier. The gas pressure was $\sim 1.5 \times 10^{-4}$ mm Hg, which was measured with an ionization gauge and was controlled by a McLeod manometer. The cross sections σ_- and σ_+ were determined from the familiar formula for single collisions.[1] The random errors of the cross sections did not exceed 15%.

3. EXPERIMENTAL RESULTS AND DISCUSSION

We determined the total cross sections σ_- for the production of free electrons, σ_+ for gas ionization, and σ_l for stripping in the collisions between fast Li, Na, K, and Cs atoms and He, Ne, Ar, Kr, and Xe atoms and H_2 and N_2 molecules. The values of σ_-, σ_+, and σ_l given in the table are the averages of three or four independent measurements, given in units of 10^{-16} cm^2, and pertain to single molecules in the cases of collisions with H_2 and N_2. We now note the characteristic features of the data.

1. As already mentioned, our measurements can be compared directly with other experimental data only for the stripping of fast lithium atoms in He, H_2, and N_2. The comparisons are shown in Fig. 1, where the dashed curves [6,7] represent values of σ_l that agree satisfactorily with the cross sections measured in [6] (especially for Li-He), but are considerably lower than those in [7]. At the lower energy limit the calorimetric detector used in [6] may have been insufficiently accurate as a means of measuring neutral atom beam intensities.

2. A comparison between the gas ionization cross section σ_+ and the alkali metal atom stripping cross section σ_l shows that in many cases σ_+ is larger than σ_l. This is a very curious result, because the ionization potential of inert ions is considerably larger than that of alkali metal atoms. The effect is probably associated with features of the electron shells of the colliding particles. The weakly bound outer electron in an alkali metal atom is quite distant from the core and may be located outside the effective interaction region when the two atoms are in close proximity. The ionization of inert gas atoms results from the collisions of relatively "rigid" systems, the core of an alkali metal atom and an inert gas atom, when considerable energy could be transferred through the statistical transfer of momentum by the electrons of filled shells. The probability of ionization can then be close to unity; this is the mechanism considered by Firsov.[8]

The situation must be essentially different when alkali metal atoms are stripped. Despite the relatively low binding energy of the outer electron there is only a low probability of energy transfer to such a "loose" system. This can be demonstrated qualitatively as follows. If we regard the outer electron approximately as a classical particle with a velocity distribution $f(v_e)dv_e$, a solution of the problem of free electron scattering yields the following relation between the energy ΔE transferred to the electron as the result of any collision and the ionization energy $U_i = \frac{1}{2} mv_0^2$:

$$\Delta E = 2mv_e v \geqslant U_i \qquad (1)$$

or

$$4\frac{v_e}{v_0}\frac{v}{v_0} \geqslant 1, \qquad (2)$$

where v_e is the orbital velocity of the electron and v is the velocity of relative motion. It is seen from (2) that for small values of v $(v/v_0 \leq 0.1)$ an outer electron can receive enough energy for its removal only when there is some probability that it has a velocity $v_e > v_0$. This probability, resulting from the tail of $f(v_e)dv_e$, is extremely small. Therefore the cross sections for alkali metal atom stripping can be smaller than the cross sections for inert gas atom ionization, especially at low collision energies.

It is interesting that the cross sections for atomic stripping exceed those for molecular ionization in the interactions between alkali metal atoms and H_2 or N_2 molecules, throughout the entire investigated energy range. The difference between σ_l and σ_+ is especially large for the collisions of the lightest particles (Li-H_2 and Na-H_2).

3. For the investigated colliding alkali metal and inert gas atoms having atomic numbers that are not very small (Z > 5) and do not differ very greatly from each other ($Z_1/Z_2 < 4$), the total cross sections for the production of free electrons (σ_-) can be compared with the total ionization cross sections calculated from Firsov's formula [8]

$$\sigma = \sigma_0 \left[\left(\frac{v}{u_0} \right)^{1/5} - 1 \right]^2 , \qquad (3)$$

in which

Colliding particles	3 keV			6 keV			10 keV			15 keV		
	σ_-	σ_+	σ_l	σ_-	σ_+	σ_l	σ_-	σ_+	σ_l	σ_-	σ_+	σ_l
Li— He	0.18	0.05	0.13	0.40	0.07	0.33	0.65	0.1	0.55	0.95	0.11	0.84
Ne	0.32	0.22	0.10	0.61	0.34	0.27	0.92	0.48	0.44	1.21	0.61	0.60
Ar	0.36	0.26	0.10	0.94	0.54	0.40	1.64	0.98	0.66	2.28	1.43	0.85
Kr	0.60	0.37	0.23	1,08	0.68	0.40	1.84	1.12	0.72	2.86	1.66	1.20
Xe	1.36	0.84	0.52	2,30	1.36	0.94	3.46	2.08	1.38	5.40	3.10	2.30
H_2	0.50	0.08	0.42	0.80	0.10	0.70	1.28	0.14	1.14	1.90	0.18	1.72
N_2	2.34	0.64	1.70	3.66	1.06	2.60	5.00	1.47	3.53	6.25	1.85	4.40
Na — He	—	—	—	0.23	0.06	0.17	0.37	0.09	0.28	0.5	0.14	0.36
Ne	0.52	—	—	0,90	0.50	0,40	1.26	0.64	0.62	1.64	0.78	0.86
Ar	0.80	0.43	0.37	1.20	0.58	0.62	1,60	0,74	0.86	2.0	0.9	1.1
Kr	0.13	0.06	0.07	0,76	0.41	0.35	1.11	0.76	0.35	1.44	1.06	0.38
Xe	0.92	0.64	0.28	1.76	0.95	0.81	2.60	1.24	1.36	3.44	1.54	1.90
H_2	—	—	—	0.47	0.09	0.38	0.70	0.12	0.58	0.96	0.14	0.82
N_2	1.83	0.96	0.87	2.70	1.27	1.43	3.55	1.53	2.02	4.40	1.74	2.66
K — He	0.13	—	—	0.30	—	—	0.51	—	—	0.72	—	—
Ne	0.31	0.27	0.04	0.56	0.34	0.22	0.84	0.41	0.43	1,17	0.50	0.67
Ar	2.07	1.22	0.85	2.91	1.68	1.23	3.74	2.16	1.58	4,50	2,56	1.94
Kr	2.55	2.0	0.55	3.58	2.6	0.98	4.45	3,11	1.34	5,10	3.45	1.65
Xe	3.40	2,40	1.0	5.0	3.3	1.7	6.3	4.0	2.30	7.30	4.56	2.84
H_2	0.38	0.07	0.31	0.61	0.15	0.46	0.91	0.27	0.64	1.23	0.37	0.86
N_2	2.23	1.13	1.10	3,78	1.54	2.24	5.0	2.0	3.0	6.13	2.4	3.73
Cs — He	0.06	—	—	0.18	0.02	0.16	0.26	0.04	0.22	0.42	0.05	0.37
Ne	0.18	0.07	0.11	0.38	0.11	0.27	0.63	0.13	0.50	0.93	0.16	0.77
Ar	1.60	0.50	1.1	2.20	0.65	1.55	2.90	0.85	2.05	3.65	1.05	2.60
Kr	2.30	1.10	1.20	3.30	1.55	1.75	4.33	2.0	2.33	5.30	2.3	3.0
Xe	3.40	1.90	1,50	4.85	2.90	1,95	6.50	3,75	2,75	7,90	4.50	3.40
H_2	0.13	0,03	0,10	0.30	0.04	0.26	0.46	0.08	0.38	0,60	0.13	0.47
N_2	1.50	0.55	0.95	2,40	0.95	1.45	3.80	1.40	2.40	5.25	1,95	3.30

Colliding particles	20 keV			25 keV			30 keV		
	σ_-	σ_+	σ_l	σ_-	σ_+	σ_l	σ_-	σ_+	σ_l
Li — He	1.23	0.14	1.09	1.52	0.17	1.35	1.80	0,18	1,62
Ne	1,52	0.71	0.81	1.75	0,82	0,93	1.95	0.97	0,98
Ar	2,90	1,73	1.17	3.52	2.02	1.50	4.12	2.20	1.92
Kr	3.82	2.08	1.74	4,73	2.36	2.37	5,60	2.66	2.94
Xe	7.15	4,08	3.07	8,50	4.80	3.70	9.93	5.60	4.33
H_2	2.33	0.23	2,10	3,06	0.26	2,80	3.42	0.29	3.13
N_2	7.10	2.22	4.88	7,87	2.60	5.27	8.40	2.90	5.50
Na — He	0.62	0.16	0.46	0.72	0.20	0.52	0.80	0.22	0.58
Ne	2.0	0.92	1.08	2.26	1.02	1.24	2.50	1.12	1.38
Ar	2,32	1.02	1.3	2.62	1,14	1.48	2.88	1,24	1.64
Kr	1,72	1,18	0.54	2.0	1.3	0.7	2.22	1.40	0.82
Xe	4.16	1,80	2.36	4.8	2.0	2.80	5,34	2.22	3.12
H_2	1.10	0.15	0,95	1.30	0.17	1.13	1.47	0.19	1.28
N_2	5.13	1.90	3.23	5,85	2.0	3.85	6,40	2.10	4.30
K — He	0.90	0.13	0,77	0.98	—	—	1,08	—	—
Ne	1.47	0,62	0.85	1.74	0.72	1.02	2.03	0.82	1.21
Ar	5.05	2.81	2.24	5.50	3.0	2.5	5.90	3.16	2.74
Kr	5.65	3.70	1,95	6.05	3.90	2.15	6.40	4.0	2.40
Xe	8.35	4.92	3.43	9.00	5.22	3.78	9.40	5,45	3.95
H_2	1,45	0.45	1.0	1.67	0,52	1.15	1.86	0,58	1.28
N_2	7.15	2.80	4.35	8.00	3.15	4.85	8,55	3.40	5.15
Cs — He	0.60	0.06	0.54	0.81	0,08	0.73	0.96	0.09	0.87
Ne	1.17	0.20	0,97	1.35	0.25	1,10	1.45	0.30	1.15
Ar	4.20	1.24	2.96	4.55	1.35	3.20	5.05	1.43	3.62
Kr	6.10	2,50	3.60	6.75	2.70	4.05	7.30	2.90	4.40
Xe	8.95	5.12	3.83	9,90	5.65	4.25	10.35	5.85	4.50
H_2	0.73	0.17	0.56	0.92	0.20	0.72	—	—	—
N_2	6.55	2.30	4.25	7,50	2,65	4.85	8.40	3.0	5.40

$$\sigma_0 = \frac{32.7 \cdot 10^{-16}}{(Z_1 + Z_2)^{2/3}}\ \mathrm{cm}^2,$$

$$u_0 = \frac{23.3 U_i}{(Z_1 + Z_2)^{5/3}} \qquad (4)$$

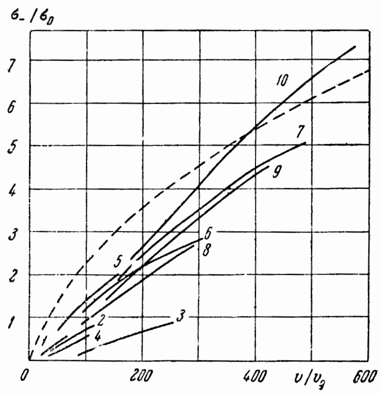

FIG. 2. Total cross sections for the production of free electrons (solid lines) measured in the present work and Firsov's theoretical curve [8] (dashed line). 1 – Na-Ne, 2 – Na-Ar, 3 – Na-Kr, 4 – K-Ne, 5 – K-Ar, 6 – K-Kr, 7 – K-Xe, 8 – Cs-Ar, 9 – Cs-Kr, 10 – Cs-Xe.

represents the characteristic cross sections and velocities, U_i is the ionization energy of the most weakly bound electron (that of the alkali metal atom in our case), Z_1 and Z_2 are the atomic numbers of the colliding particles. Figure 2 shows the ratio $\sigma_-/\sigma_0 = f(v/u_0)$ based on (3) as a universal curve (the dashed line). The solid curves represent the indicated colliding particles. The theoretical cross sections are too large, especially for the lighter atoms (Na-Ne, Ar, Kr; K-Ne). In the cases of the heavier gases the better agreement with theory is evidently associated with the increases of the measured total cross sections σ_- resulting from double ionization, which was not fully taken into account in the theory. For example, our analysis of slow ions formed in collisions between 15-keV K atoms and Xe atoms showed that Xe^{2+} constituted about 18%. We can therefore conclude that Firsov's statistical theory of ionization, which neglects the aforementioned features of the electron shells of alkali metals, is not valid for ionization processes involving the alkali metals, Firsov's theory is in considerably better agreement with experiment for collisions of inert gas atoms in inert gases. This is well shown in Fig. 3, where $\sigma_-(v)$ curves [1,2] are compared with Firsov's calculated curve.

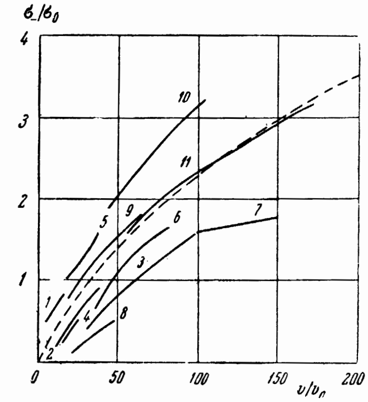

FIG. 3. Total cross sections σ_- for the production of free electrons measured in [1,2] (solid lines) and Firsov's theoretical curve [8] (dashed line). 1 – Ne-Ne, 2 – Ne-Ar, 3 – Ne-Kr, 4 – Ar-Ne, 5 – Ar-Ar, 6 – Ar-Kr, 7 – Ar-Xe, 8 – Kr-Ne, 9 – Kr-Ar, 10 – Kr-Kr, 11 – Kr-Xe.

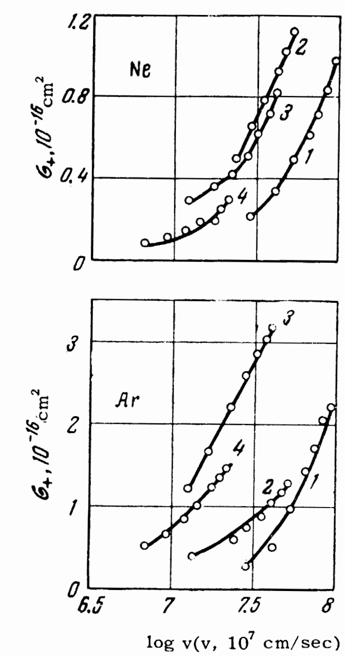

FIG. 4. Cross sections for the ionization of Ne and Ar atoms by Li, Na, K, and Cs atoms versus the logarithm of velocity. 1 – Li, 2 – Na, 3 – K, 4 – Cs.

4. In the velocity range investigated by us the ionization cross sections increase continuously with the incident particle velocity. For any given velocity v the cross sections usually increase with the atomic number of the struck particles (or with the decreasing ionization potential of the latter). Exceptions are observed only for close values of the atomic numbers (Li-He, Na-Ne, K-Ar, Cs-Xe). For example, in Fig. 4, where the ionization cross sections of neon and argon ionization by different alkali metals are compared, we find maximum values of σ_+ for identical velocities v in the cases Na-Ne and K-Ar. The stripping cross sections are also considerably larger for collisions between inert gas atoms and atoms of the adjacent alkali metal. This effect was ob-

served in [3] for Na-Ne and K-Ar at lower energies, and was even more pronounced for collisions between identical inert-gas atoms.[1,2]

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