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Multiple Ionization of Rare Gases by H^+ and He^+ Impact

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Multiple ionization of rare gases by H^+ and He^+ impact

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Absolute cross sections for multiple ionization (σ_q) of He, Ne, Ar, and Kr are presented for proton-impact energies between 10 and 4000 keV. Charge states up to 6+ are observed for Kr. The results are obtained by measuring relative yields of multiply charged ions with a time-of-flight spectrometer and normalizing to total ion-production cross sections $\sigma_T = \sum q\sigma_q$. The results are compared with existing proton- and electron-impact data. Relative multiple- to single-ionization cross-section ratios are also presented for He^+ impact.

I. INTRODUCTION

In measurements of ionization cross sections, whether they are for total electron yield[1,2] or are differential in electron emission angle and energy,[3] no ionization charge-state information is recorded. For a theoretical interpretation, it is generally assumed that single-ionization dominates. Double ionization which occurs as a result of autoionization or inner shell ionization is assumed to play a small role. This is true for electron-impact ionization.[4–12] However, it is well established from Auger[13,14] and x-ray[15,16] spectral features that fast, heavy-ion bombardment can produce multiple ionization in a single collision.

In these fast ion-atom collisions, the large nuclear to electronic mass ratio imparts most of the energy to the electrons and produces slow multiply charged target ions. These slow ions closely resemble those occurring in hot plasmas; thus there has been an active interest in their production and their interaction cross sections. In recent years, the concentration has been on very highly charged slow ion production by fast heavy-ion bombardment.[17–21] Previously, production of low to moderately charged ions by proton impact has been investigated experimentally[22–26] and theoretically.[27–31]

We present data for absolute and relative multiple ionization of noble-gas targets by 10–4000-keV H^+ and He^+ impact. Although we concentrate on the lower charge states that are predominantly produced in these collisions, we demonstrate that the relative abundance of Ar^{q+} ($q = 2$–5) produced by 2-MeV He^+ is comparable to that produced by 35-MeV Cl^{6+} impact.

EXPERIMENTAL TECHNIQUE

A. Apparatus

The relative charge-state spectrum of ions produced in the collision was measured by attaching a time-of-flight (TOF) ion charge-state analyzer to a parallel plate condenser system used for total ion- and electron-production measurements (see Fig. 1). A collimated, monoenergetic ion beam passes between the condenser plates and is collected in a biased Faraday cup. The target gas completely fills the condenser system at a pressure of ~0.5 mTorr. A transverse electric field (~25 V/cm) between the condenser plates extracts the electrons and slow ions produced and accelerates them toward the plates. Ions, initially having near-zero energy, are accelerated to an energy qV. They exit the condenser system through a small aperture and enter a field-free drift tube. Near the drift-tube entrance a set of deflector plates are normally biased to deflect the ions away from the exit aperture. By pulsing the deflection field to zero for a short time ΔT, an ion pulse can pass undeflected through the drift tube. Upon exiting the tube, ions are accelerated to the channeltron cone and are counted. Ions of different charge states travel through the tube in times proportional to $(qV/M)^{-1/2}$ where M is the ion mass.

A TAC (time-to-amplitude converter) module, started by the deflector pulse and stopped by the arrival of an ion at the channeltron, produces an ion time-of-flight spectrum as shown in Fig. 2. Owing to the relationship be-

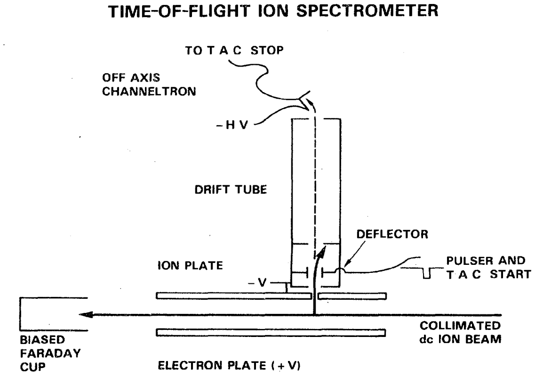

FIG. 1. Time-of-flight ion analyzer showing ion beam passing between condenser plates. Extracted ions normally follow curved trajectory (solid line) but can be pulsed through the drift tube (dashed curve) when deflection field is zero.

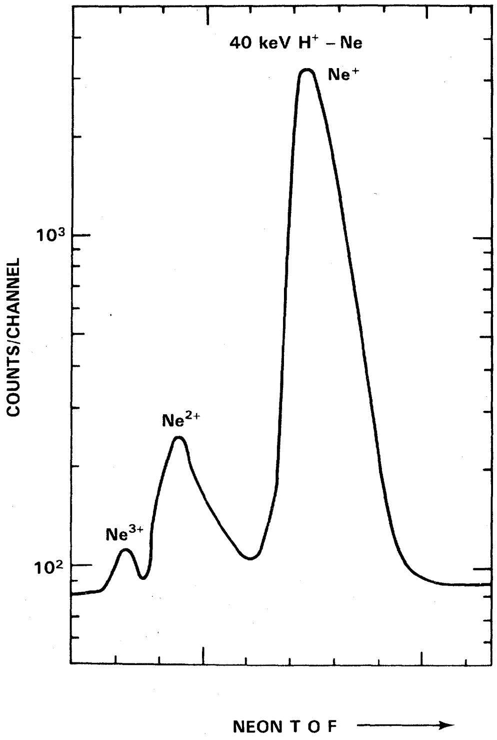

FIG. 2. Time-of-flight charge-state spectrum for slow multiply charged neon ions produced by 40-keV H^+ impact.

tween ion flight time and charge state, these spectra can be easily interpreted for atomic targets. Relative multiple ionization cross sections are then obtained by subtracting a linear background and integrating the various peaks. These relative cross sections can be put on an absolute scale by normalizing to our total ion production cross sections,[2] where

$$\sigma_T = \sum_q q\sigma_q = \sigma_1 \sum_q R_q \ , \qquad (1)$$

where σ_T is the total ion-production cross section, σ_q is the cross sections for charge state q, and $R_q = \sigma_q / \sigma_1$ is the relative yield of charge state q.

EXPERIMENTAL TESTS

Charge-state ratios $R_2 = [Ne^{2+}]/[Ne^+]$ were measured for H^+-Ne collisions for a variety of experimental conditions. It was found that as the extraction field was increased from 2 to 65 V/cm, the Ne^+ and Ne^{2+} signals increased and then remained constant as the field strength was further increased to 150 V/cm. This is consistent with our observations of the total ion signal (see Ref. 2). The ratio R_2 was found, however, to be constant within $\pm 10\%$ throughout this entire range. We therefore chose to use an operating value of approximately 25 V/cm since this provided better charge-state resolution due to the smaller potential gradient across the beam width and hence a better defined ion energy in the drift tube.

The detection efficiency of the channeltron was tested as a function of ion energy by varying the cone potential from -3200 to -500 V while keeping the gain constant. Using Ar^{q+} ions, R_2 and R_3 were measured and found to be constant within 10% for channeltron cone voltages ranging from -3200 to -1400 V. All data were accumulated using a cone voltage of -3200 V.

The largest experimental uncertainty is due to a charge-dependent ion transmission through the TOF spectrometer. This is because of the finite ion flight time between the deflectors. Since ions are transmitted only when the deflectors field is zero, ions having longer flight times (i.e., lower charges and thus lower accelerated energies) will be transmitted less efficiently. This transmission function can be shown to be of the form

$$\tau_q = 1 - \frac{T_f}{\Delta T} \ , \qquad (2)$$

where T_f is the ion flight time in the deflection field, ΔT is the zero-field pulse time, and T_f is given by $l/\sqrt{2qV/M}$, where l is the effective deflector length. Assuming unit detector efficiency, and no other parameters affecting ion transmission through the drift tube, the number of ions detected (N) is then given by

$$N = N_0 \frac{\Delta T}{T} \tau_q$$
$$= N_0 \frac{\Delta T}{T} \left[1 - \frac{1}{\Delta T} \frac{l}{\sqrt{2qV/M}} \right] ,$$

where N_0 is the number of ions entering the flight tube and T is the pulse repetition time.

Measurements of Ne^+, Ne^{2+}, and Ne^{2+}/Ne^+ as a function of pulse width ΔT (Fig. 3) clearly demonstrate this

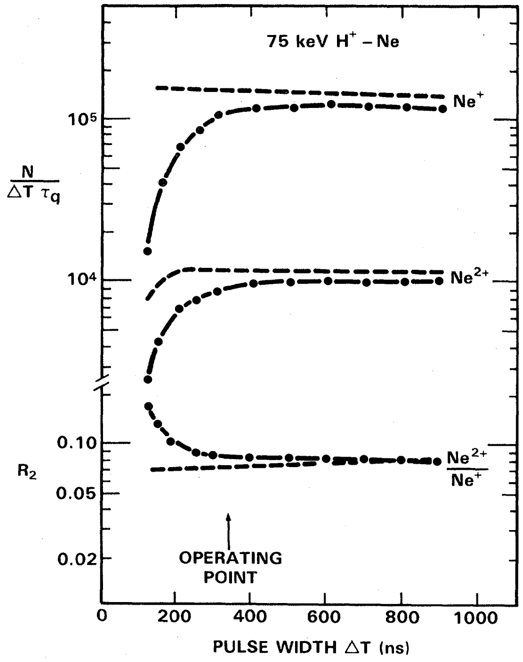

FIG. 3. Yields of Ne^+, Ne^{2+}, and Ne^{2+}/Ne^+ as a function of deflection pulse width (ΔT). Solid curves are drawn through the raw data points. Dashed curves are obtained after correction for transmission effects as described in the text.

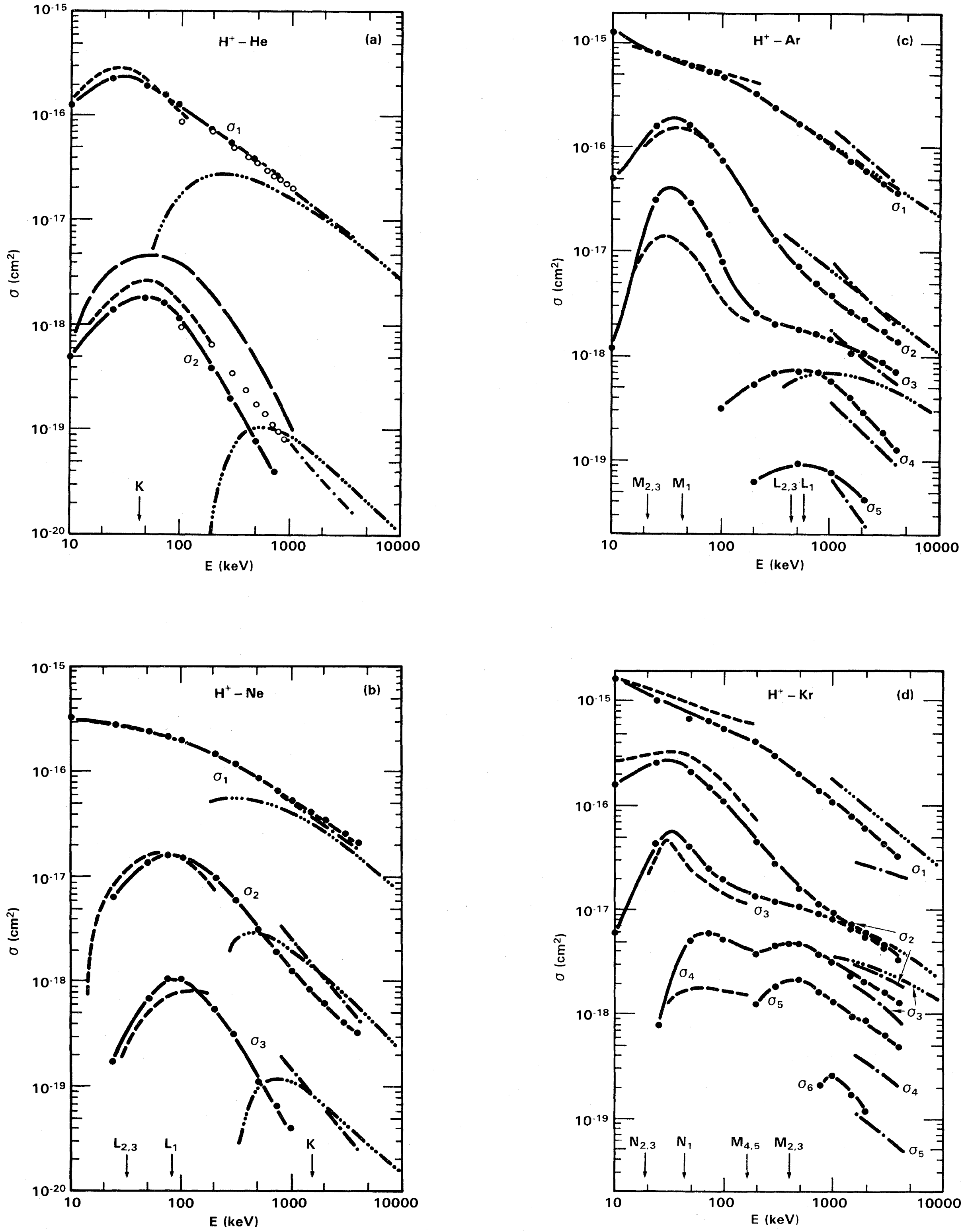

FIG. 4. Total and multiply charged ion-production cross sections for H$^+$-He, Ne, Ar, and Kr collisions. For H$^+$ impact: ——●——, σ_q (present data); — — —, σ_q (Ref. 23); ——·——, σ_q (Ref. 22); ○, σ_q (Ref. 25); —— ——, σ_2, theory for He^{2+} (Ref. 27). For e^- impact (scaled velocity): ——. . .——, σ_q (combined data from Refs. 5 and 9—12). The vertical arrows indicate proton velocities that match the bound target-electron velocities.

TABLE I. Cross sections of multiple ionization of H^+ impact (σ_n in 10^{-16} cm^2).

		E(keV)													
Target	n	10	25	50	75	100	200	300	500	750	1000	1500	2000	3000	4000
He	1	1.28	2.37	1.95	1.50	1.25	0.762	0.566	0.378	0.274					
	2	5.02^{-3}	0.0148	0.0184	0.0166	0.0173	3.93^{-3}	1.98^{-3}	8.96^{-4}	3.92^{-4}					
Ne	1	3.39	2.88	2.45	2.20	2.07	1.53	1.23	0.893	0.699	0.573	0.432	0.347	0.257	0.211
	2		0.0637	0.139	0.158	0.152	0.0998	0.0616	0.0316	0.0194	0.0131	8.90^{-3}	6.56^{-3}	4.11^{-3}	3.32^{-3}
	3		1.67^{-3}	6.96^{-3}	0.0107	0.0111	5.55^{-3}	3.37^{-3}	1.11^{-3}	4.25^{-4}					
Ar	1	13.5	7.85	6.32	5.33	5.00	3.39	2.54	1.77	1.37	1.05	0.738	0.587	0.470	0.386
	2	0.486	1.60	1.64	1.07	0.775	0.258	0.135	0.0726	0.0507	0.0389	0.0273	0.0235	0.0179	0.0139
	3	0.0102	0.318	0.297	0.144	0.0800	0.0254	0.0198	0.0174	0.0163	0.0147	0.0103	0.0106	8.93^{-3}	7.14^{-3}
	4					3.00^{-3}	5.22^{-3}	6.86^{-3}	6.91^{-3}	7.13^{-3}	5.99^{-3}	4.21^{-3}	2.94^{-3}	1.88^{-3}	1.27^{-3}
	5						6.10^{-4}		9.19^{-4}		7.74^{-4}		4.37^{-4}		
Kr	1	16.7	10.3	6.84	6.47	5.76	4.06	3.01	2.4	1.44	1.10	0.803	0.622	0.442	0.330
	2	1.57	2.65	2.12	1.52	1.12	0.459	0.279	0.161	0.115	0.0931	0.0729	0.0613	0.0491	0.0334
	3	0.0622	0.435	0.403	0.247	0.194	0.136	0.121	0.111	0.0938	0.0823	0.0661	0.0570	0.0464	0.0354
	4		8.13^{-3}	0.0526	0.0605	0.0522	0.0377	0.0470	0.0487	0.0386	0.0326	0.0237	0.0208	0.0174	0.0131
	5						0.0126	0.0184	0.0218	0.0163	0.0135	0.00963	0.00871	0.00623	0.00492
	6									2.18^{-3}	2.64^{-3}	1.70^{-3}	1.21^{-3}		

transmission effect. For small pulse widths the Ne^+ intensity falls by an order of magnitude, the Ne^{2+} by a lesser amount, and hence their ratio R_2 increases by a factor of 2.

The effective deflector length l and hence the transmission τ_q were obtained from $N/\Delta T$ vs $1/\Delta T$ plots for various charge states of Ne and Ar. The transmission corrections, when applied to the data in Fig. 3, produce Ne^+, Ne^{2+}, and Ne^{2+}/Ne^+ yields that are constant to within 10% as shown. Experimentally, one would like to use a large pulse width ΔT to minimize this correction term but is limited to a smaller ΔT because of charge resolution requirements. Hence compromise values of (τ_{+q}/τ_{+1}) ranging from ~ 0.9 for $q=2$ to ~ 0.7 for $q=6$ were used.

Consideration must also be given to resonant charge transfer as the ions travel through the target gas from the interaction region to the detector (~ 10 cm). Using published data[32] for charge transfer in slow Ar^{q+}-Ar collisions and assuming similar cross sections for other systems under investigation here, we calculated less than 1% alteration of charge states for ions traveling from the interaction region to the channeltron.

Combining all the experimental uncertainties we estimate that our relative multiple ionization ratios range in accuracy from $\pm 15\%$ for R_2 to as much as $\pm 50\%$ for the highest charge states measured. The increased errors for higher charge states are due to resolution limitations, application of larger transmission corrections, and poorer peak to background statistics. Normalizing to our absolute total ion-production cross sections, which are accurate to better than 10%, introduces only small additonal errors.

RESULTS

In Fig. 4 we present partial ionization cross sections for proton impact on He, Ne, Ar, and Kr along with previously published data. Total ion-production cross sections σ_T obtained from Ref. 2 were used to obtain the absolute partial ionization cross sections shown and tabulated in Table I. For the lighter targets He and Ne, the total ion-production results essentially from single ionization of the target; with multiple ionization becoming increasingly more important for the heavier targets. In the case of H^+-Kr, approximately 50% of the total ion-production cross section is through multiple ionization events. The observed ionization could result from direct Coulomb interaction between the projectile and target or from electron capture by the projectile from the target. Since electron-capture cross sections are maximum when the proton velocity matches that of the bound electrons, we have indicated these conditions for the various target shells[33] with arrows. Electron capture from the K shell contributes strongly to single and double ionization of He. Capture from the L shell of Ne can produce up to triply charged Ne. For the heavier targets both outer- and inner-shell electron-capture effects can be observed. For example, up to triply ionized Ar is associated with M-shell electron capture whereas at higher energies L-shell capture produces Ar^{3+}, Ar^{4+}, and Ar^{5+}. In krypton, outer-shell

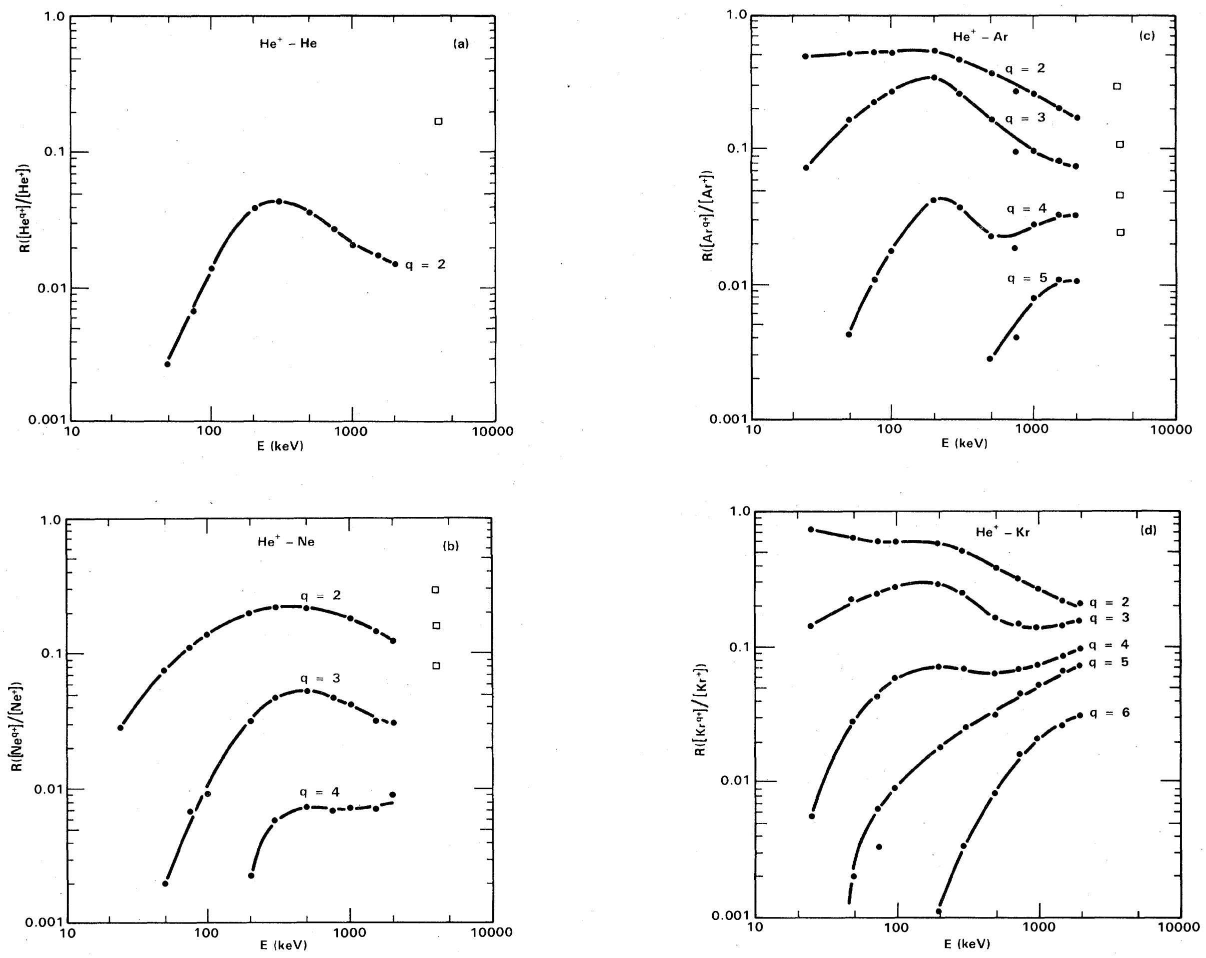

FIG. 5. Ratios of multiply to singly charged ion production in He$^+$-He, Ne, Ar, and Kr collisions. ——●—— (present data); □ (ratios obtained for 35-MeV Cl^{6+} impact, Ref. 17).

capture produces up to four-times ionized Kr, whereas capture from the *M* shell is important for producing Kr^{3+}-Kr^{6+}. Capture from inner shells probably produces these higher charge states through Auger decays and cascading processes. Capture from the outer shells must be associated with simultaneous additional outer-shell ionization to produce the doubly and triply charged ions that we observe at lower energies. In addition to these processes, direct multiple outer-shell ionization is possible.

In Fig. 4 we also compare our results with previously measured multiple ionization cross sections by proton impact. The data of Solov'ev *et al.*[23] (dashed curve) covers the energy range from 10 to 180 keV and, except for the cases of Ar^{3+} and Kr^{4+}, agrees quite well with the present results. For those two cases our cross sections are larger but have approximately the same energy dependence. Between 40 and 500 keV, Werner[24] has recently measured multiple ionization ratios associated with simultaneous electron production. We are unable to compare with his data since he could only observe ion production when an electron is also liberated in the collision. Thus the singly charged ions that he observed were produced only by direct ionization processes while the multiply charged ions could result from direct ionization as well as single-electron capture. At MeV energies, the data of Wexler[22] (dotted-dashed curve) generally disagree with the present results except for single ionization of He, Ne, and Ar. The data by Puckett and Martin[25] (○'s) agree for single ionization of He but are larger for He^{2+} production. The only theoretical values shown[27] are for double ionization (long dashed curve) of He which shows approximately the same energy dependence as the present data but overestimates the cross sections.

The data for proton impact are also compared with equivalent velocity electron-impact results. Because of the large amount of electron-impact data the dotted curves shown are a combination of several experimental investigations.[5,9–12] At higher energies, there is excellent agreement between the single-ionization cross sections measured for e^- and H$^+$ impact on He,Ne,Ar. For multiple ionization the electron-impact cross sections are larger than those for proton impact. One would expect that since the charge-exchange channel is closed for electron impact, proton impact would cause more multiple ioniza-

TABLE II. Relative multiple- to single-ionization cross-section ratios for He^+ impact (σ_n/σ_1 in %).

Target	n	E (keV) 25	50	75	100	200	300	500	750	1000	1500	2000
He	2		0.263	0.645	1.36	3.81	4.28	3.59	2.66	2.00	1.72	1.48
Ne	2	2.78	7.51	11.1	13.3	19.9	22.1	21.3	19.8	18.1	14.6	12.6
	3		0.202	0.670	0.910	3.13	4.75	5.27	4.65	4.21	3.21	3.06
	4					0.227	0.599	0.740	0.678	0.711	0.716	0.914
Ar	2	50.0	51.4	53.4	51.4	55.6	47.5	37.6	27.0	26.0	20.4	17.3
	3	7.38	16.9	22.5	26.9	35.1	26.4	16.6	9.71	9.74	8.24	7.64
	4		0.425	1.08	1.78	4.23	3.68	2.22	1.87	2.84	3.31	3.26
	5							0.286	0.419	0.800	1.10	1.06
Kr	2	73.8	64.6	59.5	60.1	59.0	50.8	38.0	31.2	26.5	21.7	20.7
	3	14.2	22.9	24.3	27.8	29.3	24.1	16.1	14.8	13.8	14.2	15.3
	4	0.563	2.84	4.28	5.99	6.98	6.90	6.19	6.92	7.22	8.63	9.53
	5		0.203	0.628	0.890	1.80	2.53	3.16	4.53	5.25	6.53	7.23
	6					0.110	0.330	0.830	1.64	2.12	2.63	3.16

tion. However, in a recent letter McGuire[34] discusses this problem for ionization of He and shows that the opposite is true. The present results demonstrate the same effect for other gases. The results for e^-,H^+-Kr are contradictory to the above, possibly because of enhanced H^+-impact cross sections due to M-shell electron capture. In the data presented, it must be remembered that certain ionic charge states of interest cannot be distinguished from background impurity ions, e.g., $H_2{}^+$ from He^{2+}; $N_2{}^+$ and CO^+ from Kr^{3+}; and N^+, $N_2{}^{2+}$, $O_2{}^{2+}$ from Kr^{6+} and Ar^{3+}. These contaminant ions could result because of insufficiently low background gas pressures or contamination of the target-gas supply system. No background gas contributions were observed for the present experimental setup, and although contamination of the target gas could not be investigated, we have no reason to suspect this form of contamination for the present data. Commercially available gases of 99.9% purity or better were used for all of our measurements.

In Fig. 5 and Table II we present our relative cross sections for producing multiply charged ions in He^+-He, Ne, Ar, and Kr collisions. Multiple to single charge-state production ratios are given since no reliable total charge-state production cross sections were available for the energies and targets of interest. Although no direct comparison is shown, we find strongly enhanced multiple charge production as compared to proton impact. This large degree of multiple ionization (note in particular He^+-Kr at 2 MeV) can be quite important in evaluating total ionization data where only total charge is recorded. A comparison with 35-MeV Cl^{6+} multiple charge-state yields[17] is made to demonstrate that relative Ar^{q+} ($2<q<5$) production is almost as effective using 2-MeV He^+ beams as for heavier beams emerging from much larger accelerators. However, as pointed out in Ref. 18 the overall cross sections for producing highly charged ions increase rapidly with increased projectile charge state. But He^+ beams can be useful for studies involving low to moderately charged ions.

SUMMARY

We have presented absolute and relative cross sections for multiply charged ion production by H^+ and He^+ impact on He, Ne, Ar, and Kr. The data, obtained by TOF ion charge-state analysis agree well with the previous data of Solov'ev *et al.*[23] but generally disagree with the high-energy data of Wexler.[22] For Ar and Kr targets, it was shown that multiple ionization is an important part of the total ion-production cross sections which could play a significant role in the analysis of differential or total ionization data. For He^+ impact multiple ionization is still more important. We plan to continue this investigation of multiple charge-state production due to direct ionization and charge transfer in the near future.

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[1] J. W. Hooper, D. S. Harmer, D. W. Martin, and E. W. McDaniel, Phys. Rev. 125, 2000 (1961).

[2] M. E. Rudd *et al.* (unpublished).

[3] L. H. Toburen, *High Energy Ion-Atom Collisions,* in Proceedings of the International Seminar on High-Energy Ion-Atom Collision Processes (Debrecen, Hungary, 1981), edited by D. Berenyi and G. Hock (Elsevier, New York, 1982), Vol. 2, p. 53.

[4] M. J. Van der Wiel, Th. M El-Sherbini, and L. Vriens, Physica (Utrecht) 42, 411 (1969).

[5] K. Stephan, H. Helm, and T. D. Märk, J. Chem. Phys. 73, 3763 (1980).

[6] V. Schmidt, N. Sandner, and H. Kuntzemuller, Phys. Rev. A 23, 1743 (1976).

[7] F. A. Stuber, J. Chem. Phys. 42, 2639 (1965).

[8] R. E. Fox, J. Chem. Phys. 33, 200 (1960).

[9] B. Adamczyk *et al.*, J. Chem. Phys. 44, 4640 (1966).

[10] P. Nagy, A. Skutlartz, and V. Schmidt, J. Phys. B 13, 1249 (1980).

[11] B. L. Schram, A. J. H. Boerbeem, and J. Kistemaker, Physica (Utrecht) 32, 185 (1966).

[12] A. Gaudin and R. Hagemann, J. Chem. Phys. 64, 1209 (1967).

[13] N. Stolterfoht, F. J. de Heer, and J. Van Eck, Phys. Rev. Lett. 30, 1159 (1973).

[14] N. Stolterfoht, D. Schneider, R. Mann, and F. Folkman, J. Phys. B 10, L282 (1977).

[15] M. D. Brown *et al.* Phys. Rev. A 9, 1470 (1974).

[16] H. Oona, Phys. Rev. Lett. 32, 571 (1974).

[17] C. L. Cocke, Phys. Rev. A 20, 749 (1979).

[18] T. J. Gray, C. L. Cocke, and E. Justiniano, Phys. Rev. A 22, 849 (1980).

[19] H. F. Beyer, K.-H. Schartner, and F. Folkman, J. Phys. B 13, 2459 (1980).

[20] A. S. Schlachter *et al.*, Phys. Rev. A 23, 2331 (1981).

[21] P. Hvelplund, H. K. Haugen, and H. Knudsen, Phys. Rev. A 22, 1930 (1980).

[22] S. Wexler, J. Chem. Phys. 41, 1714 (1964); 44, 2221 (1966).

[23] E. S. Solov'ev *et al.*, Zh. Eksp. Teor. Fiz. 42, 659 (1962) [Sov. Phys.—JETP 15, 459 (1962)].

[24] H.-C. Werner, Ph.D. thesis, Free University, Berlin 1982 (unpublished).

[25] L. J. Puckett and D. W. Martin, Phys. Rev. A 1, 1432 (1970).

[26] E. Horsdal Pedersen and L. Larsen, J. Phys. B 12, 4085 (1979).

[27] A. K. Kaminsky and M. I. Popova, J. Phys. B 9, L177 (1976).

[28] A. Kumar and B. N. Roy, Can. J. Phys. 56, 1255 (1978).

[29] A. Salop, Phys. Rev. A 9, 2946 (1974).

[30] A. Langeberg, *Physics of Electronic and Atomic Collisions,* Proceedings of the Ninth International Conference on the Physics of Electronic and Atomic Collisions, Seattle, 1975, edited by J. S. Risley and R. Geballe (Washington University Press, Washington, 1975), p. 902.

[31] Th. M. El-Sherbini and M. J. Van der Wiel, Physica (Utrecht) 62, 19 (1972).

[32] C. L. Cocke *et al.*, Phys. Rev. Lett. 46, 1671 (1981).

[33] J. A. Bearden and A. F. Burr, Rev. Mod. Phys. 39, 125 (1967).

[34] J. H. McGuire, Phys. Rev. Lett. 49, 1153 (1982).