# PROTON-INDUCED K-SHELL HOLE PRODUCTION ON Ar AND Kr (\*)

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ABSTRACT — Proton-induced K-shell X-ray production cross sections for Ar and Kr are measured in the incident energy ranges 0.135 - 2 MeV and 0.5 - 2 MeV, respectively, using ion implanted targets. The corresponding ionisation cross sections are compared to other experimental results, when available, and to theoretical cross sections calculated in accordance with the PWBA, SCA and BEA models, taking into account effects due to retardation of the projectile by the nuclear Coulomb field of the target, electronic binding energy and polarisation effects and, for Kr, to electronic relativistic effects. Best overall agreement is found with the PWBA corrected values; the agreement is poor for Kr at the lowest energies used, the theoretical cross sections being roughly 50% above the experimental ones.

### 1-INTRODUCTION

There is only scant experimental information on the protoninduced K-shell ionisation cross sections of argon and krypton; the published results [1-6] are summarised in the tables of Rutledge and Watson [7] and Gardner and Gray [8]. The measurements have been made on gas targets under single collision conditions with the exception of the work of Lennard and Mitchell [3] who used Kr implanted on Si for their measurements of the Kr cross sections.

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In this paper we report on measurements of the Ar and Kr K-shell hole production cross sections using targets made by ion implantation on Be and compare our results to the PWBA [9-11] and SCA [12-13] theories corrected for electron binding and polarization effects, projectile Coulomb retardation and detlection and electronic relativistic effects; a comparison to the Coulomb effect corrected BEA theory [14-15] is also made.

# 2-EXPERIMENTAL PROCEDURE

Proton beams from 0.135 to 2 MeV were produced by the 2 MV. Van de Graaff accelerator at Sacavem; the  $H_3^+$  beam was used to obtain the 0.135 MeV incident energy while the other energies below 0.4 MeV were obtained with the  $H_2^+$  beam. The experimental set up has been described previously [16] and only a few specific points are referred here.

The Ar X-rays were detected in a Si(Li) detector with a resolution of 200 eV for Ar and the Kr X-rays in a HP Ge detector with a resolution of 250 eV for Kr. The X-ray detector was placed at an angle of 105° to the beam and accepted radiation within a solid angle of 0.548 msr. X-ray background with a pure Be foil in the target position was negligible for the integrated currents used in each run. The count rate was kept low but where appropriate dead time corrections have been made. X-ray absorption corrections have been made as described previously [16]. The efficiency of the Si(Li) detector for the Ar X-rays has been found following the method proposed by Rosner et al. [17] and using Ca SO<sub>4</sub>.

A silicon surface barrier detector of 13 keV resolution for 1 MeV protons was used to detect backscattered protons within a solid angle of 2.12 msr at 165° degrees to the beam axis. Proton spectra were taken in a multichannel analyser and the peak corresponding to the element under study was integrated to be used in the X-ray production cross section determination. Background counts, coming mainly from the Be (p,  $\alpha$ ) reaction were at each incident energy subtracted from the proton scattering spectra; at high incident energies an aluminium foil 0.025 mm thick was used in front of the particle detector to stop the alpha particles. A particle spectrum of the Ar target is shown superimposed on a background spectrum (taken with a Be foil with an equal amount of deposited carbon) in Figure 1, where the corres-

ponding difference spectrum, giving a well defined peak, is also displayed; these spectra were taken at 0.2 MeV, thus representing the worst case situation. (The Ar X-ray production cross section at 0.135MeV was obtained assuming the target thickness determined at an higher energy and measuring the incident charge.) Dead time corrections had to be made but generally did not exceed 10%; they were based on the simultaneous measurement of a peak produced by pulses fed by a mains triggered pulse generator into the particle preamplifier.



Fig. 1 — Spectra of backscattered protons at 0.2 MeV incident energy. *a*) Spectrum given by the Ar implanted on Be target superimposed on the background spectrum given by a Be foil with the same amount of deposited carbon. *b*) Difference spectrum showing clearly the peak due to the Ar atoms.

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Except for Ar at 0.135 MeV, the X-ray production cross sections were determined using the formula

$$\sigma_{\rm X}({\rm E}_i) = 4 \pi \sigma_{\rm R}({\rm E}_i, \theta) . (n_{\rm X} \Omega_n / n_n \Omega_{\rm X})$$

where  $\sigma_{R}(E_{i}, \theta)$  is the Rutherford cross section at energy  $E_{i}$  and detection angle  $\theta$ ,  $n_{X}$  and  $n_{p}$  are the corrected numbers (see below) of simultaneously detected X-rays and protons, and  $\Omega_{p}$  and  $\Omega_{X}$  are the solid angles subtended by the proton and X-ray detectors, respectively. The number  $n_{X}$  has been corrected for the detection system overall efficiency and for dead time. The number  $n_{p}$  has been corrected for dead time and for the  $E^{-2}$  dependence of the Rutherfordscattering law at incident energies where the fact that the implanted ions are below the Be foil surface makes this correction significant.

The targets were implanted in flat Be foils at 80 keV bombarding energy at ISKP, University of Bonn. We used the work of Santry and Werner [18] to obtain the ranges of the implanted ions, finding the values 17  $\mu$ gcm<sup>-2</sup> for Ar and 8  $\mu$ gcm<sup>-2</sup> for Kr, with an error estimate of  $\pm 25\%$ . The targets were positioned at 45° to the beam; thus, assuming negligible range straggling, the proton beam had to pass through Be layers 24  $\mu$ gcm<sup>-2</sup> and 11.3  $\mu$ gcm<sup>-2</sup> thick before hitting the Ar and Kr atoms, respectively. At low energies the results have to be corrected for the significant beam energy loss thus produced. The proton stopping power has been obtained from the tables of Northcliffe and Shilling [19]; the energy loss for a 24  $\mu$ gcm<sup>-2</sup> Be layer at 0.135 MeV bombarding energy is calculated to be 16.2 keV, and at 1 MeV, 5.5 keV. To obtain the X-ray production cross sections at the stated bombarding energy we assumed that the cross section varied as the  $\alpha$  power of the energy,

$$\sigma_{X}(E) = (E/(E-\Delta E))\alpha^{+2} \cdot \sigma_{X}(E-\Delta E)$$

where E is the bombarding energy and  $\Delta E$  the energy loss; the term 2 in the exponent comes from the Rutherford scattering law. The value of  $\alpha$  has been derived from the corrected theoretical PWBA cross sections. Thus to obtain the cross section value for Ar at 0.135 MeV, the measured value has to be multiplied by 2.04; at 0.2 MeV by 1.54; at 0.5 MeV by 1.09. The corresponding factor for Kr at 0.5 MeV is 1.04. The error in the cross section coming

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from a 25% error on the assumed thickness of the Be layer in front of the implanted Ar decreases with increasing incident energy and is roughly 30% at 0.135 MeV, 12% at 0.2 MeV and 6% at 0.3 MeV. The Ar and Kr layers were, of course, very thin, with a density of roughly  $5 \times 10^{-16}$  atoms cm<sup>-2</sup>.

## 3-RESULTS AND DISCUSSION

The results obtained in the present work are shown in Table I together with other available experimental values. For Kr it is seen that our results are almost a factor of 2 smaller than the gas target results of Winters et al [2] while being in agreement to within 10-20% with those obtained with a Kr implanted on Si target by Lennard and Mitchel [3]. For Ar all the previous results have been obtained with gas targets and it is seen that there is agreement with the present values to within roughly 10%, except for the value at 2 MeV which is about 25 % lower than some of the previous results [5,6].

A comparison of the present data with theoretical values derived from the PWBA, SCA and BEA theories is now made. The PWBA values were calculated following the procedures and using the tables of Basbas et al. [11]. Corrections for electron binding and polarisation effects, and for retardation and deflection of the projectile in the Coulomb field of the nucleus are included in the calculated values; electronic relativistic corrections made according to formula 4 of Amundsen et al [20] were also included in the theoretical values for Kr. In Figure 2 is plotted the ratio  $\sigma_{th}/\sigma_{exp}$  for both Ar and Kr against the logarithm of the proton incident energy represented in terms of the scaled energy variable  $y = \eta_{\rm K} / (\zeta_{\rm K} \theta_{\rm K})^2$  defined by Basbas et al [11], where  $\eta_{\rm K} = (v_1 / v_{2\rm K})^2$  is the square of the projectile velocity in units of the K-shell electron velocity,  $\theta_{\rm K}$  is the screening parameter given by the ratio of the binding energy to that of a pure hydrogenic system and  $\zeta_{\kappa}$  is a  $v_1$ -dependent function related to the binding and polarisation effects. For Ar, our incident energy range corresponds to values 0.024 < y < 0.45; for Kr, to values 0.020 < y < 0.085. It is seen that over their common y-range the ratios  $\sigma_{\rm th}/\sigma_{\rm exp}$  for Ar and Kr have their main differences for low values of y, say y < 0.03, the calculated PWBA cross sections being, relatively to the experimental ones, larger for Kr than for Ar. In this low energy range use of the Kocbach Coulomb correction

#### TABLE I

K-shell ionisation cross sections (in barn). The integers in brackets indicate powers of 10. Estimated absolute errors are  $\pm$  10 % above 0.3 MeV,  $\pm$  15 % below 0.3 MeV and  $\pm$  35 % at 0.135 MeV; corresponding estimated relative errors are  $\pm$  5 %,  $\pm$  10 % and  $\pm$  20 %, respectively. Fluorescence coefficients are taken to be 0.122 and 0.660 for Ar and Kr, respectively.

Ei	Argon		Krypton	
	This work	Previous work	This work	Previous work
2.0	3.41 (3)	3.75 <sup><i>a</i></sup> ,4.29 <sup><i>b</i></sup> ,4.00(3) <sup><i>c</i></sup>	1.20 (1)	$2.00^{a}$ , $1.00(1)^{f}$
1.9	3.57 (3)		1.03 (1)	
1.8	3.33 (3)		9.28 (0)	
1.7	3.00 (3)		8.02 (0)	
1.6	2.77 (3)		6.80 (0)	$6.20 (0)^g$
1.5	2.37 (3)	$2.61(3)^a$	5.13 (0)	$9.44(0)^a$
1.4	2.11 (3)		4.53 (0)	0.11 (0)
1.3	1.89 (3)		3.22(0)	$3.41(0)^{g}$
1.2	1.65 (3)		2.43 (0)	
1.1	1.39 (3)		1.78 (0)	
1.0	1.21 (3)	$1.15(3)^d$	1.45 (0)	$1.51(0)^{g}$
0.9	1.04 (3)	$9.67(2)^d$	1.03 (0)	9.48(-1)a
0.8	8.16 (2)	$7.66(2)^d$	6.68(-1)	$7.36 (-1)^g$
0.7	5.73 (2)	$5.61(2)^d$	3.77(-1)	
0.6	3.94 (2)	$3.93(2)^d$	1.72 (-1)	
0.50	2.46(2)	$2.46(2)^d$	9.08(-2)	$1.00 (-1)^{f}$
0.45	1.84 (2)			
0.40	1.26 (2)	$(1,30)(2)^d$		
0.35	7.63 (1)			
0.30	4.75 (1)	$5.34(1)^d$		1.
0.25	2.64 (1)	$2.93 (1)^d$		10000
0.20	1.29 (1)	$1.30(1)^d$		
0.135	3.41 (0)	$3.49(0)^{e}$		1

a: Ref. 2; b: Ref. 5; c: Ref. 6; d: Ref. 4; e: Ref. 1; f: Refs. 8, 3. g: Interpolated from the data of Refs. 8, 3.

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factor [21] for Kr would slightly improve the agreement with experiment. For Ar the introduction of the polarisation correction [11], an additive correction increasing with energy in this scaled energy range, made a significant improvement in the theory by increasing the calculated value at 2 MeV by about 20% relatively to the low energy values. Thus the more recent paper of Basbas et al [11] gives an appreciably better agreement with experiment than the older one [10].



Fig. 2 — Ratio of theoretical to experimental proton-induced ionisation cross sections,  $\sigma_{th}/\sigma_{exp}$ , for *a*) krypton and *b*) argon. The theoretical cross sections correspond to the PWBA, SCA and BEA models and are calculated as described in the text. The abcissa is the logarithm of the scaled energy variable  $y = \eta_K / (\zeta_K \theta_K)^2$  referred to in the text.

The formulae and procedures presented by Laegsgaard et al [13] were used to obtain the SCA cross sections taking into account the effects due to electron binding, Coulomb retardation and, in the case of Kr, the relativistic form of the electron wavefunctions at small distances from the nucleus. The SCA cross sections thus calculated do not allow for polarisation effects which, in the present case, are most important for Ar at the higher end of the incident energy range;

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thus one would expect the SCA cross sections for Ar to become significantly smaller than the experimental ones at those energies, as Figure 2 indeed shows. For Kr, the SCA cross sections, like the PWBA ones, are above the experimental values; this is especially so at low scaled incident energies, say y < 0.03, where the Coulomb and relativistic corrections are large. One may note here that there is fair agreent between the present results for Kr and those obtained by Lennard and Mitchell [3,8].

In summary, one may say that the corrected PWBA theory gives the best overall fit to the present results (within about  $\pm$  10% of the average value of the ratio  $\sigma_{\rm th} / \sigma_{\rm exp}$  for Ar) and that the inclusion of the polarisation effect provided a significant improvement in the case of Ar. For low values of the scaled energy variable y the fit is better for Ar than for Kr. The binding energy correction procedure gives, of course, similar correction factors for Ar and Kr and, indeed, seems very accurate if a comparison is made between the results of Ford et al [22] using self consistent electron wavefunctions in a second order Born calculation and those obtained following Basbas et al [11]. Thus, the fact that at low energies the Ar results are better fitted than those of Kr possibly indicates that the procedures to take into account nuclear Coulomb effects and electronic relativistic effects (which are much smaller for Ar than for Kr) still need to be improved with the net effect of reducing the calculated cross section at low values of the scaled energy variable.

The ratio  $k_{\beta}/k_{\alpha}$  for Kr was measured at 1.8 MeV incident proton energy with the results  $k_{\beta}/k_{\alpha} = 0.178 \pm 0.005$ . This ratio has been determined in proton-induced ionisation by other authors with the results  $0.189 \pm 0.010$  [2] and  $0.167 \pm 0.009$  [3]. Multiple ionisation induced by proton impact is expected to be small in this Z-region [23] and this is reflected in the good agreement of the present experimental result with the value  $k_{\beta}/k_{\alpha} = 0.173$  calculated by Scofield [24] under the assumption that the excited atom is singly ionised.

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#### REFERENCES

- [1] HARRISON, K. G., TAWARA, H., HEER, F. J., Physica 66, 16 (1973).
- [2] WINTERS, L. M., MACDONALD, J. R., BROWN, M. D., ELLS-WORTH, L. D., CHIAO, T., Phys. Rev., A7, 1276 (1973).
- [3] LENNARD, W. N., MITCHELL, I. V., Phys. Rev., A12, 1723 (1975).
- [4] LANGENBERG, A., VAN ECK, J., J. Phys., B9, 2421 (1976).
- [5] RANDALL, R. R., BEDNAR, J. A., CURNUTTE, B., COCKE, C. L., Phys. Rev., A13, 204 (1976).
- [6] PRESSER, G., private communication.
- [7] RUTLEDGE, C. H., WATSON, R. L., At. Data and Nucl. Data Tables, 12, 195 (1973).
- [8] GARDNER, R. K., GRAY, T. J., At. Data and Nucl. Data Tables, 21, 515 (1978).
- [9] MERZBACHER, E., LEWIS, H. W., Handbuch der Physik (ed. by S. Flugge), Vol. 34, p. 166, Berlin-Gottingen-Heidelberg: Springer (1958).
- [10] BASBAS, G., BRANDT, W., LAUBERT, R., Phys. Rev., A7, 983 (1973).
- [11] BASBAS, G., BRANDT, W., LAUBERT, R., Phys. Rev., A17, 1655 (1978).
- [12] BANG, J., HANSTEEN, J. M., K. Dan. Videnk. Selsk. Mat. Fys. Medd., 31, 1 (1959).
- [13] LAEGSGAARD, E., ANDERSEN, J. U., LUND, M., Proc. 10th Intern. Conf. on the Physics of Electronic and Atomic Collisions (ed. by G. Watel), p. 353, North-Holland Publ. Co. (1978).
- [14] MCGUIRE, J. H., RICHARD, P., Phys. Rev., A8, 1374 (1973).
- [15] HANSEN, J. S., Phys. Rev., A8, 822 (1973).
- [16] LOPES, J. S., JESUS, A. P., FERREIRA, G. P., GIL, F. B., J. Phys., B11, 2181 (1979).
- [17] ROSNER, B., GUR. D., SHABASON, L., Nucl. Inst. and Meth., 131, 81 (1975).
- [18] SANTRY, D. C., WERNER, R. D., Nucl. Inst. and Meth., **139**, 135 (1976).
- [19] NORTHCLIFFE, L. C., SCHILLING, R. F., Nucl. Data Tables, A7, 233 (1970).
- [20] AMUNDSEN, P. A., KOCBACH, L., HANSTEEN, J. M., J. Phys., B9, L203 (1976).

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- [21] KOCBACH, L., Phys. Norvegica, 8, 187 (1976).
- [22] FORD, A. L., FITCHARD, E., READING, J. F., Phys. Rev., A16, 133 (1977).
- [23] SEE e.g. RICHARD, P., Atomic Inner-Shell Processes (ed. by B. Crasemann), p. 73, New York, Academic Press (1975).
- [24] SCOFIELD, J. H., Atomic Inner-Shell Processes (ed. by B. Crasemann), p. 265, New York, Academic Press (1975).