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INNER SHELL IONIZATION OF NOBLE
GAS ATOMS BY PROTON BOMBARDMENT

by

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TABLE OF CONTENTS

Introduction	1
Background	1
Theory	4
Fluorescence Yields	18
Experimental Apparatus and Methods	24
Apparatus	24
Corrections Applied to X-ray Yields	51
Data Analysis	70
Results and Discussion	84
Appendix I	87
Appendix II	104
List of Symbols	109
References	111
Acknowledgments	113

INTRODUCTION

Background

Collisions of heavy, charged particles with atoms at keV energies and greater are, by nature, highly inelastic. Inelastic energy losses which are large enough to suggest the production of inner shell vacancies in the colliding particles have been reported by several workers. A survey of this work has been given by Kessel¹. Kessel, et.al.² have observed energy losses so large--almost 30 keV in large angle I^{3+} on Xe collisions--that they can be accounted for only in terms of vacancy production in the L and M shells of the colliding particles. Thus, the study of inner shell vacancy production is a means of gaining more insight into the dynamics of the collision process. In particular, an understanding of the coupling between elastic and inelastic processes in these collisions has yet to be attained.

An atom that contains an inner shell vacancy can de-excite either radiatively by the emission of an x-ray photon or non-radiatively by Auger and Coster-Kronig processes with the resultant emission of electrons. Therefore, inner shell ionization can be studied indirectly by the observation of these photons or electrons or both. Spectroscopic analysis of the Auger electrons is a relatively new technique and has proven quite successful^{3,4}. The emphasis of this paper, however, will be on the observation of the characteristic x-rays.

Merzbacher and Lewis⁵ have given a review of the early experiments in which characteristic x-ray production was studied. Since their article was published, a number of other experiments have been performed to measure inner shell ionization cross sections in bombardment of solid targets with

protons⁶⁻¹⁴. As far as proton-induced ionization of gaseous targets is concerned, however, little work has been done outside of just identifying the characteristic x-rays². Saris and Onderdelinden¹⁵ have measured cross sections for the production of L-shell x-rays of argon bombarded by 70 to 130 keV protons; however, their work overlaps the present experiment neither in incident proton energy nor in the energy of the emitted x-rays. Specifically, the aim of this experiment is to study K-shell ionization of argon and krypton and L-shell ionization of krypton by observing the characteristic x-rays produced in collisions of these gases with 1.5 to 4.5 MeV protons. A summary of this work has been submitted to the International Conference on Inner Shell Ionization Phenomena and will be published in the proceedings of that conference. In addition, the summary is included as Appendix I of this paper. Also included as an appendix (Appendix II) are the initial results on characteristic x-ray production in proton bombardment of xenon.

Although the technical difficulties associated with using gas targets are greater than for solid targets, the former affords a simpler means of data analysis and a greater amount of information about the collision process than the latter. There are two reasons for this: 1) The targets can be made thin enough that self-absorption is negligible, and 2) single collision conditions prevail.

In the case of solid targets, the thick target yield of x-rays per unit solid angle in the direction of the detector is given by⁵

$$I_{\mu}(x_0) = (n/4\pi) \int_0^{x_0} e^{-\mu(x_0 - x)} \sigma_e[E(x)] dx$$

where $\sigma_e[E(x)]$ is the cross section per atom per incident proton for emission of an x-ray, E is the incident proton energy, x is the residual

range of the proton with a total range of x_0 , n is the number of target atoms per unit volume, and μ is the absorption coefficient of the target for its own characteristic x-radiation. It should be noted that the form of the exponential in the above formula applies only if the distance traversed by an x-ray in the target is the same as the distance traversed by the proton that produced it. If negligible self-absorption is assumed, the exponential drops out so that one has

$$I(x_0) = \frac{n}{4\pi} \int_0^{x_0} \sigma_e[E(x)] dx.$$

For single collision conditions, each proton strikes at most a single target atom so that the dependence of $\sigma_e[E(x)]$ on x drops out, and the total range of protons is simply L , the interaction length viewed by the detector. Then for monoenergetic protons incident on a sufficiently thin target, the x-ray yield reduces to

$$I = \frac{nL}{4\pi} \sigma_e(E). \quad (1.1)$$

Thus, a knowledge of stopping powers and absorption coefficients is unnecessary for obtaining x-ray production cross sections in thin target experiments.

The greatest advantage in using thin targets, however, is the amount of information that can be obtained. In thick targets, the incident ion may undergo many collisions both before and after the collision that produces a given x-ray. Thus, one does not know either the initial or final state of the ion. Moreover, information regarding scattering of the ion is lost. Such information is quite important if one is to obtain useful results regarding the collision process. X-rays alone cannot give these

results, and herein lies the usefulness of the thin target. The single collision criterion means that it is possible, at least in principle, to correlate the production of the x-ray with such characteristics of the collision as impact parameter, charge state, elastic and inelastic energy loss, excitation state, and lifetimes. It is hoped that the present experiment, in addition to providing experimental data on x-rays in a region where such data has not previously been available, will also be the first step in a study of heavy-ion collisions by particle-photon coincidence methods.

Theory

One may speak qualitatively of the process of inner shell ionization by heavy, charged particle impact in terms of the velocities of the collision partners. Letting v be the velocity of the incident proton and u be the effective velocity of the atomic electron, then the three regions to consider are $v^2 \gg u^2$, $v^2 \sim u^2$, and $v^2 \ll u^2$. In the region of $v^2 \gg u^2$, large energy transfers from the incident proton to the electron are possible so that one would expect inner shell ionization to occur. The behavior of the cross section here is analogous to the situation for electron excitation of atoms, an approximate $1/E$ dependence being exhibited in both cases¹⁶. Moreover, the stopping cross section in this velocity region is dominated by a $1/E$ dependence¹⁷ and is a result primarily of the electronic component. The general trend of decreasing ionization cross section with increasing energy in the high energy region is a result of the decreasing time of interaction.

In the adiabatic region of $v^2 \ll u^2$, the situation is quite different for proton- and electron-induced ionization. For the electron-atom collision, the indistinguishability of the incident and atomic electron results in an

energy threshold below which ionization cannot occur. This threshold energy is simply equal to the binding energy of the atomic electron¹⁸. For proton bombardment, however, this complication does not arise. In this case, the increased mass of the projectile coupled with the high velocity of the inner shell electrons allows energy transfers large enough to eject inner shell electrons. One can obtain a rough qualitative view of this by using conservation of energy and momentum to determine the maximum energy which a heavy particle of mass M and velocity v can transfer to a light particle of mass m and velocity u in a free collision. This energy transfer is

$$T_m = \left[\frac{2Mm}{(M+m)^2} \right] (Mv - mu)(v+u). \quad 5' \quad (1.2)$$

For the proton-electron collision we have $(m/M) \ll (v/u)$ so that (1.2) reduces to $2mv(v+u)$, showing that for a given value of v , an increase in u means a larger energy transfer. Thus, in the adiabatic region, the ionization cross section increases with increasing proton energy. The steepness of ascent should depend on the binding energy of the atomic electron, the cross section rising less rapidly for more tightly bound electrons. Therefore, for a given atom and a given projectile energy, the cross section for K-shell ionization will be lower than that for L-shell ionization in the adiabatic region.

For the region $v^2 \sim u^2$, equation (1.2) reduces to a value on the order of the binding energy of the electron. The cross section for ionization is expected to have its maximum in this region.

Merzbacher and Lewis⁵ have used the Born approximation to derive a form for the cross section for inner shell ionization by proton and alpha particles in the region $v \lesssim u$. The perturbation that results in ionization

is taken to be the Coulomb interaction between colliding particles so that the cross section is proportional to the square of the matrix element

$$\int \psi_f^*(\underline{R}, \underline{r}) \frac{ze^2}{|\underline{R} - \underline{r}|} \psi_i(\underline{R}, \underline{r}) d\underline{r} d\underline{R}.$$

ψ_i and ψ_f are the initial and final wave functions of the system, ze is the projectile charge, and $|\underline{R} - \underline{r}|$ is the separation of the particles.

A number of assumptions were employed⁵ in order to simplify the problem. These are summarized below.

- 1) Setting $u = 0$ in equation (1.2) and letting I_s be the binding energy of the electron in its orbit, the following restriction is placed on the maximum energy transfer:

$$T_m \approx (4m/M)E \gg I_s \quad (1.3)$$

where the symbol $\gg$ means not much greater than. This is equivalent to assuming that $v \sim u$ and results in a simplification of the form factor in the calculations.

- 2) Letting Z be the nuclear charge of the atom and s the principal quantum number, one imposes

$$s(zu/Zv) \ll 1. \quad (1.4)$$

This condition, which is equivalent to $ze^2/hv \ll 1$, places a lower limit on the incident particle energy and also on the atomic number of the target. It also serves to motivate assumptions 3) to 5).

- 3) The distortion of the wave function of the projectile by the electron is ignored. This assumption should begin to break down for the slowest collisions.

- 4) The wave function of the electron is taken to be that for the

unperturbed atom. This means that polarization of the electron orbit by the projectile is neglected. This should be valid for small projectile charge ($z \sim 1$ and $z \ll Z$).

5) The Coulomb repulsion between the projectile and the nucleus is ignored by using plane waves for projectile wave functions. Such an approach may be valid if the distance of closest approach, b , of the projectile to the nucleus is small compared with the radius, a_s , of the electron orbit. Explicitly,

$$b/a_s \ll 1.$$

where $b = zZe^2/E$. This condition is essentially the same as (1.4); in fact, Henneberg has demonstrated the validity of the plane wave approximation for K-shell ionization if (1.4) is true¹⁹.

6) Relativistic effects associated with the velocity of the atomic electron are neglected. This assumption should get worse as the atomic number of the target increases and should be worse for the K-shell than for the L-shell.

Proceeding on the above assumptions, the low-energy limit of the K-shell ionization cross section was obtained. In this derivation, atomic screening was ignored, hydrogenic wave functions were used for the electron, and an S-wave approximation was employed with the result that

$$\sigma_K = (2^{20} \pi a_0^2 / 45) (z^2 \gamma_K^4 / Z^4) \quad (1.5)$$

where σ_K is the total K-shell ionization cross section and γ_K is the dimensionless energy parameter mE/MZ^2 (Rydberg). Thus, in the adiabatic region σ_K increases as the fourth power of E and decreases as the twelfth power of Z .

Atomic screening was taken into account by introducing an effective nuclear charge Z_s and a screening number θ_s that may vary between 0 and 1. Z_s reflects the reduction in Z due to screening by inner electrons, whereas θ_s reflects the reduction in binding energy due to screening by outer electrons. θ_s is defined by

$$I_s = \theta_s(Z_s^2/s^2)(\text{Rydberg}) \quad (1.6)$$

where I_s is the actual ionization potential of the shell with principal quantum number s .

These effects were included in a calculation of the cross section for ionization of the s -shell to obtain a result that is applicable over the entire energy region of interest ($szu/Z \ll v \sim u$). The result is

$$\sigma_s = (8\pi a_0^2 z^2 / Z_s^2 \gamma_s) f_s(\theta_s, \gamma_s) \quad (1.7)$$

where f_s can be determined from a knowledge of the form factor. Form factors have been given for the K, L and M shells and subshells^{5,20}, and tables of f_s have been compiled by Khandelwal, et.al.²¹ for the K shell and L subshells.

To obtain theoretical cross sections involving the particular parameters of this experiment, appropriate substitutions were made into formula (1.7). Values of Z_s were determined using Slater's rules²² with the result that $Z_K = Z - 0.3$ and $Z_L = Z - 4.15$. Ionization potentials for computing θ_s were taken from Bearden and Burr²³, and values of f_s were obtained by interpolating on the aforementioned tables²¹. Since proton projectiles were used throughout the experiment, $z = 1$. The parameters Z_s , I_s , and θ_s are summarized in Table I.

It is convenient to plot $Z_s^4 \sigma_s / z^2$ versus γ_s for a given θ_s . This has

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TABLE I

Born Approximation Parameters for Argon and Krypton

Atomic Level	Z	Z_s	$I_s(\text{ev})$	θ_s
Ar K	18	17.7	3202.9	.752
Kr K	36	35.7	14325.6	.826
Kr L _I	36	31.85	1921.0	.557
Kr L _{II}	36	31.85	1727.2	.501
Kr L _{III}	36	31.85	1674.9	.486

been done for K-shell ionization of argon and krypton and L-shell ionization of krypton. The results are presented in Plates I and II. The general shape of the curves is as expected. The cross section rises sharply from zero to a broad maximum and then falls off slowly. The maximum lies at a smaller value of γ_s for σ_L than for σ_K , this being consistent with the fact that the ionization potential of the K-shell is greater than that of the L-shell for a given atom. A similar, although much smaller effect is seen for the L subshells of krypton for which a downward trend of the maximum with γ_L occurs as one progresses from the most tightly bound L_I shell to the least tightly bound L_{III} shell. In all cases, the maximum occurs, as predicted, in the region of T_m slightly greater than I_s . This is easily seen from the following approximate expression for γ_s :

$$\gamma_s = T_m / 4s^2 I_s$$

The magnitude of the cross section is also a function of the binding of the electron. Ionization of the L-shell of krypton is three orders of magnitude greater than that of the K-shell of the same atom, the latter being two orders of magnitude less than that of the K-shell of argon. The difference in ionization of the L_I and L_{II} subshells of krypton is similarly attributable to binding, whereas the large difference in the L_{II} and L_{III} cross sections is the result of the doubled occupancy of the L_{III} subshell over the L_{II} subshell.

A final note concerns the energy region (1.5 to 4.5 MeV) of this experiment. The theory under discussion predicts that the maximum in the L-shell ionization cross section lies in this region. The experimental determination of the slope of the excitation function in the region of the broad maximum will then provide a test of the theory. For argon K-shell

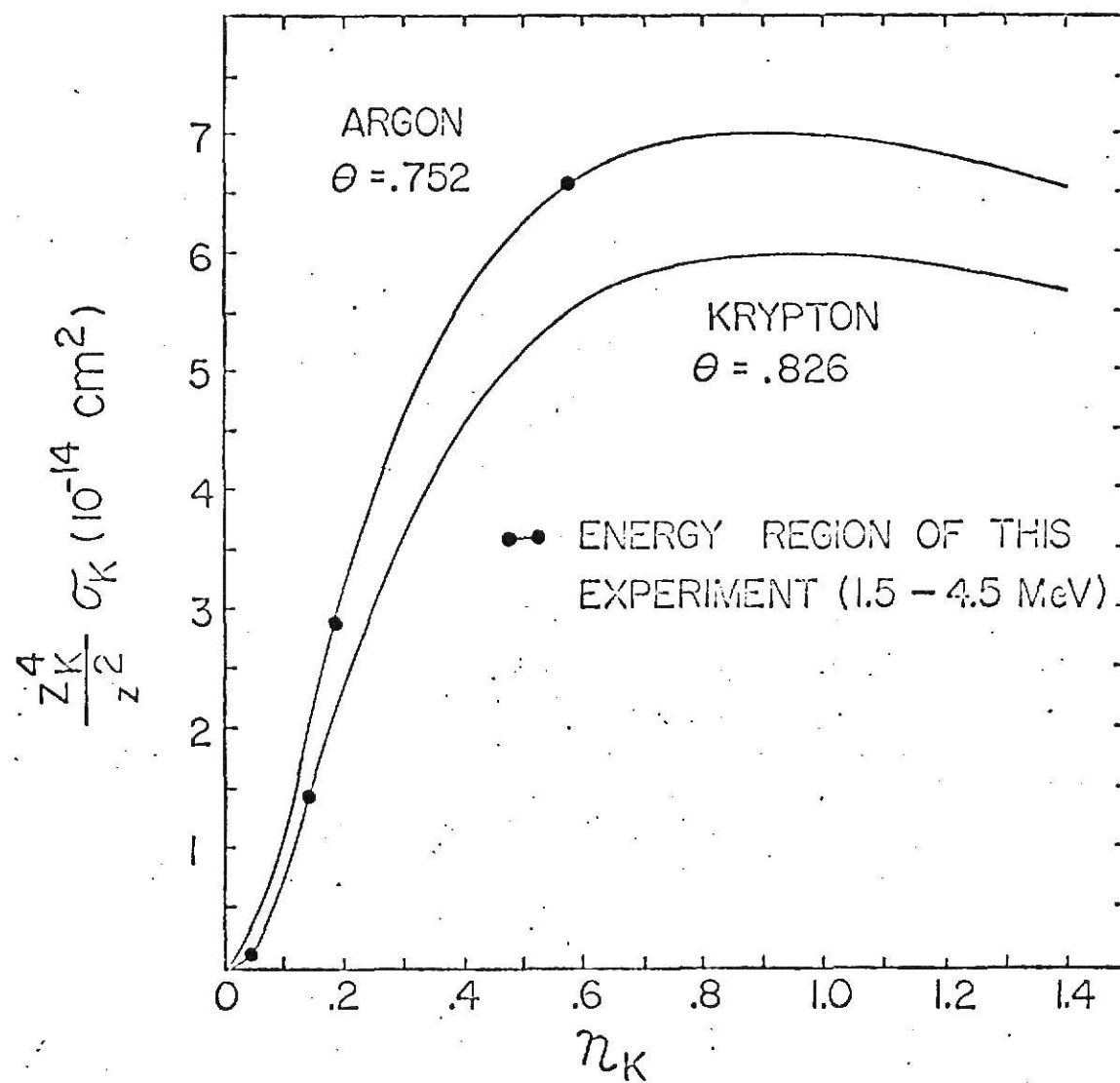
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EXPLANATION OF PLATE I

Theoretical ionization cross sections for the K-shell of argon and krypton in the Born approximation⁵. The abscissa is the dimensionless energy parameter $\gamma_K = mE/MZ_K^2(\text{Rydberg})$, and the cross section is scaled by Z_K^4/z^2 . θ is the screening parameter $I_K/Z_K^2(\text{Rydberg})$. The energy region of this experiment is enclosed by dark circles for both the argon and krypton curves.

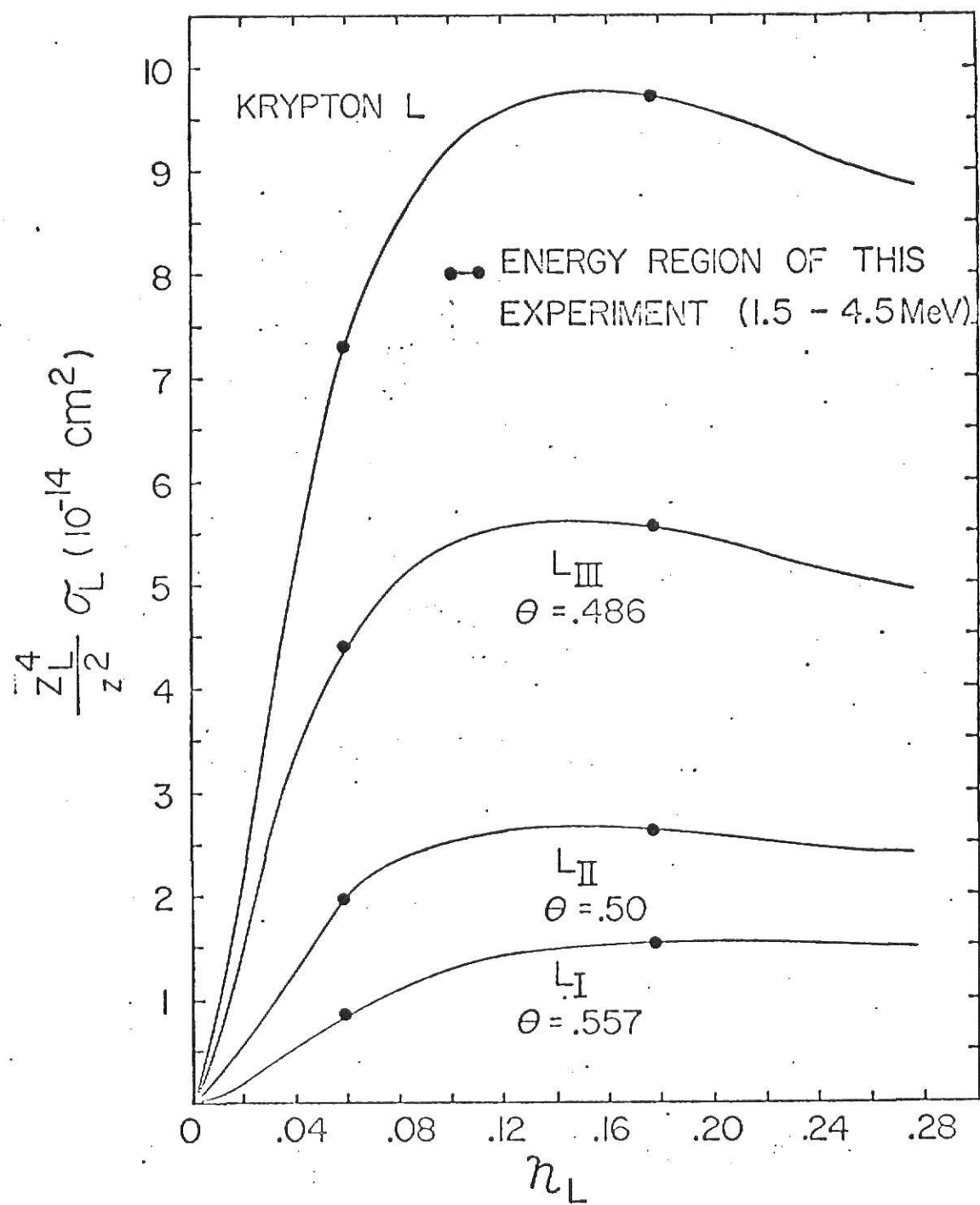
PLATE I



EXPLANATION OF PLATE II

Theoretical ionization cross sections for the L-shell of krypton in the Born approximation⁵. The total L-shell cross sections as well as the subshell cross sections L_I , L_{II} , and L_{III} are given. The abscissa is the dimensionless energy parameter $\mathcal{N}_L = mE/MZ_L^2$ (Rydberg), and the cross section is scaled by Z_L^4/z^2 . θ is the screening parameter $4I_L/Z_L^2$ (Rydberg). The energy region of this experiment is enclosed by the dark circles.

PLATE II



ionization the experimental energy region is on the rising edge of the peak; whereas, for the krypton K-shell, it is much nearer the adiabatic region.

Many experiments have been performed to determine the validity of the Born approximation in the region of low incident proton velocities⁸⁻¹¹. In general it has been found that below incident energies of about 500 keV, the experimental data lie below the theoretical predictions, the disagreement being worst for the lowest energies. At these energies, differences between theory and experiment of an order of magnitude or more are typical. It has been suggested by all of the above authors that the discrepancy is due, at least in part, to Coulomb deflection of the incoming particle by the target nucleus. As a result of this Coulomb interaction, the projectile cannot penetrate the inner shells of the target atom so deeply, thus causing a reduction in the cross section. This implies a breakdown of assumption 5) of the Merzbacher treatment.

Bang and Hansteen²⁴ have approached the problem of low-energy collisions semi-classically, assuming a hyperbolic path for the projectile. Khan, et.al.²⁵ have demonstrated that this treatment brings the theoretical curve closer to the experimental data for K-shell ionization of aluminum by protons of energy between 20 and 200 keV, but considerable discrepancies still remained. Brandt and Laubert^{11,26} have been able to account for this remaining discrepancy by including in the cross section a correction for the binding of the atomic electron to the projectile, an effect neglected (assumption 4) by the Born approximation treatment of Merzbacher.

At the high-energy end of the interval included in this treatment; that is, at energies in the vicinity of the peak of the excitation function, the theory for K-shell ionization has been found to predict results lower than

the experimental data. The data of Bissinger, et.al.¹² for K-shell ionization of calcium, titanium, and nickel by 2 to 28 MeV protons is approximately 25% to 50% higher than theoretical predictions at the peak of the excitation function and beyond; however, agreement improves at lower energies. Watson, et.al.²⁷ have made similar observations for 30 to 80 MeV alpha particles incident on various solid targets.

These experimental results may reflect the breakdown of condition (1.3) in addition to that of condition (1.4) for the lighter elements and the neglect of relativistic effects for the heavier elements. The inclusion of relativistic considerations has the effect of increasing the theoretical cross sections because of the increased electron density at the origin. Jamnik and Zupancic²⁸ have performed relativistic calculations of the K-shell ionization cross section by using Dirac Coulomb wave functions for the atomic electron. Choi²⁹ has done similar calculations for L-shell ionization. Little has been done in terms of evaluating these results for particular elements.

In view of the departure from theory exhibited by both the low-energy and high-energy data, one would expect to find that the experimental excitation function crosses the theoretical one on the low-energy side of the peak. Such a crossover point has been observed by Hart, et.al.¹⁰ for proton bombardment of oxygen and by Khan, et.al.²⁵ for proton bombardment of aluminum. The location of this point, of course, will depend on the choice of fluorescence yield, a subject to be discussed in the following section.

Recently, Garcia³⁰ has proposed a classical, binary-encounter model to describe inner shell ionization at the higher proton energies. In addition, he has presented an approximate correction to account for nuclear repulsion of the proton at lower incident energies. The binary-encounter model employs the assumption that a direct energy exchange between the incident

proton and a single atomic electron is the major interaction in producing inner shell ionization. The differential scattering cross section, in the laboratory frame, for this energy exchange is integrated over all possible energy exchanges, with the lower limit being the electron binding energy and the upper limit being the proton energy. This result is then averaged over the velocity distribution of the electron to obtain the total cross section. Garcia shows that this approach yields the same result as the quantum-mechanical impulse approximation and does account adequately for the ionization process for values of $E/I_s \gtrsim 300$. At high energies, the binary-encounter (impulse approximation) results are in fair agreement with the Born approximation⁵, and at low energies, the former results corrected for nuclear repulsion of the proton agree quite well with the calculations of Bang and Hansteen²⁴.

An interesting and useful result of Garcia's treatment is that the product of the ionization cross section for a particular shell and the square of the binding energy of an electron in that shell is a function only of the ratio of incident proton energy to electron binding energy. There is no dependence on other target parameters, such as nuclear charge, as long as nuclear repulsion of the proton is negligible. This means that a plot of $I_s^2 \sigma_s$ against E/I_s should be a universal function for all target atoms. Garcia³¹ has compared this function with existing proton data for K-shell ionization and has found quite good agreement at high energies. Likewise, Bissinger, et.al.¹² have obtained a universal curve for the Born approximation by plotting $\sigma_K Z_K^4$ against γ_K/θ_K^2 and have compared it with proton data in the same energy region. They find that in nearly all cases the data lie above the curve. Therefore, the impulse approximation appears to be better than the Born approximation at describing the inner shell ionization

process for high energies. The universal curves for both approximations are presented in Plate III. For the Born approximation, the ordinate and abscissa have been modified to $\sigma_K Z_K^4 (\text{Rydberg})^2 \sigma_K = I_K^2 \sigma_K$ and $M \gamma_K / m \theta_K = E / I_K$.

Fluorescence Yields

Since the filling of an inner shell vacancy can proceed either by radiative or non-radiative means, an experiment which looks only at the x-rays produced does not directly provide ionization cross sections. To convert from an x-ray production cross section to an ionization cross section, a knowledge of the fluorescence yield and, in some cases, Coster-Kronig yields is necessary. The fluorescence yield, ω , of a particular shell (or subshell) is defined in terms of the total x-ray transition rate, T_x , and the total Auger transition rate, T_a , of that shell (subshell):

$$\omega = T_x / (T_x + T_a). \quad (1.8)$$

It should be emphasized here that an Auger transition is one in which a vacancy in a particular shell is filled by an electron from a higher shell with the resultant ejection of a second electron from the atom to conserve energy and angular momentum. The Auger yield of the shell (subshell) is similarly

$$a = T_a / (T_x + T_a).$$

For the K-shell, Auger and x-ray transitions are the only ones which can occur so that one has

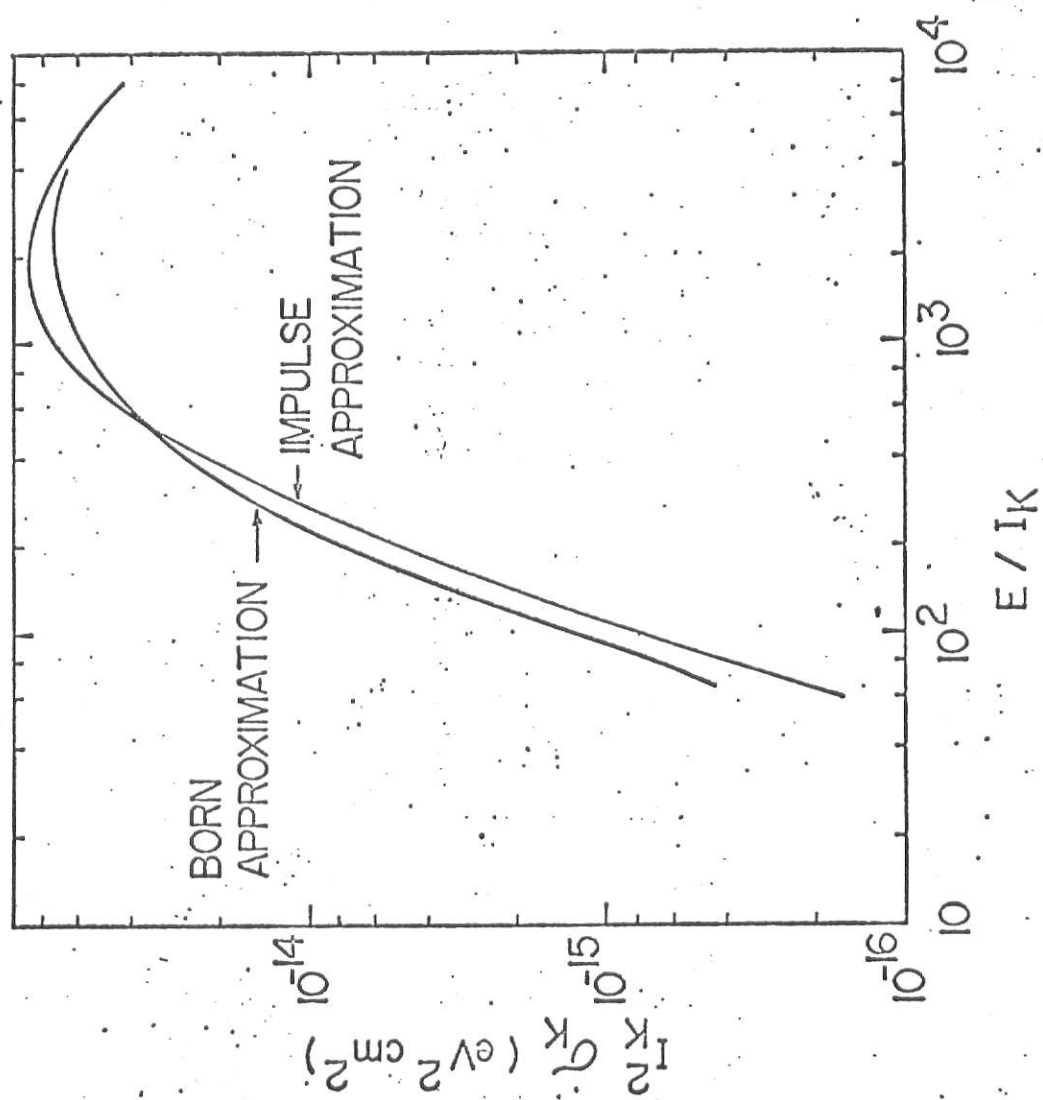
$$\omega_K + a_K = 1.$$

ω_K relates the x-ray production cross section σ_{ke} to the ionization cross

EXPLANATION OF PLATE III

Universal curves for K-shell ionization as a function of the scaled energy parameter E/I_K . The ordinate is the scaled K-shell ionization cross section $I_K^2 \sigma_K$. The two curves are the Born approximation result of Merzbacher and Lewis⁵ and the binary-encounter result of Garcia³¹.

PLATE III



section σ_K by the simple relation

$$\sigma_K = \sigma_{Ke} / \omega_K \quad (1.9)$$

For the L-shell and higher shells, however, the situation is not quite so simple. Here, one has the possibility of Coster-Kronig transitions occurring. Such a transition involves the filling of a vacancy in a particular subshell by an electron from the same shell but higher subshell with the resultant ejection of an electron from a higher shell. These transitions become important because they have transition rates on the order of those for Auger processes. They have the effect of redistributing initial vacancies to other subshells before an x-ray is emitted and thus effectively increasing the fluorescence yield. For example, an L_1 (L_I) vacancy may be shifted to the L_2 (L_{II}) or L_3 (L_{III}) subshell where the probability of de-excitation by x-ray emission is greater. To account for such effects, it is necessary to redefine the fluorescence yield in terms of probabilities for Coster-Kronig transitions. Let f_{ij} be the probability that the Coster-Kronig transition $L_i \rightarrow L_j$ occurs. Then the new definition of "fluorescence yield", γ_i , for subshell L_i becomes

$$\begin{aligned} \gamma_1 &= \omega_1 + f_{12}\omega_2 + (f_{13} + f_{12}f_{23})\omega_3, \\ \gamma_2 &= \omega_2 + f_{23}\omega_3, \\ \gamma_3 &= \omega_3. \end{aligned} \quad (1.10)$$

Now one has the following relations among the a_i , ω_i , and f_{ij} for the L-shell:

$$\begin{aligned} \omega_1 + a_1 + f_{12} + f_{13} &= 1, \\ \omega_2 + a_2 + f_{23} &= 1, \\ \omega_3 + a_3 &= 1. \end{aligned}$$

The relation (1.9) then becomes

$$\nu_1 \sigma_{L_1} + \nu_2 \sigma_{L_2} + \nu_3 \sigma_{L_3} = \sigma_{Le} \quad (1.11)$$

This relation demands that a comparison of theory with experiment be done by first converting theoretical subshell ionization cross sections to a total L-shell x-ray cross section. Or equivalently, one can first convert the experimental x-ray cross section to a total L-shell ionization cross section by use of the relation

$$\sigma_L = \sigma_{Le} / \bar{\nu}_L \quad (1.12)$$

where $\bar{\nu}_L$ is a mean L-shell fluorescence yield obtained by weighting the ν_i with their relative occupation numbers, N_i , and assuming that the ionization probability is the same for electrons in each subshell. This results in

$$\begin{aligned} \bar{\nu}_L &= N_1 \nu_1 + N_2 \nu_2 + N_3 \nu_3 \\ \text{or } \bar{\nu}_L &= \frac{1}{4}(\nu_1 + \nu_2) + \frac{1}{2} \nu_3. \end{aligned} \quad (1.13)$$

This last result is valid only for a closed L-shell as is the case for argon and krypton. A similar development may be carried out for higher shells but that is not a concern of this experiment.

Values of fluorescence yields used in this experiment were taken from several sources. For argon, the value of $\omega_K = .119 \pm .007$ was obtained from the measurements of Bailey and Swedlund³², while the value of $\omega_K = .675$ for krypton was selected from theoretical calculations of Walters and Bhalla³³. The argon value agrees well with recent theoretical calculations³³⁻³⁵ as does the krypton value with older experimental measurements summarized by Fink, et.al.³⁶ For the L-shell of krypton, the experimental data is quite sparse. Fink lists two mean values which are quite at variance with recent Hartree-

Fock calculations³⁷⁻³⁹. The results of these calculations, which give fluorescence yields smaller than the experimental values by factors of one-third and one-sixth, were used in this experiment. Values were selected from reference 37 since this reference gives a complete set of fluorescence and Coster-Kronig yields for the krypton L-shell. These yields are listed below. The mean L-shell fluorescence yield, $\bar{\nu}_L$, calculated from these values is .0238.

$\omega_1 = .00185$	$\nu_1 = .0240$	$f_{12} = .230$
$\omega_2 = .0220$	$\nu_2 = .0241$	$f_{13} = .686$
$\omega_3 = .0236$	$\nu_3 = .0236$	$f_{23} = .0897$

Obviously, ionization cross sections will be sensitive to the choice of fluorescence yield. This is particularly true for the L-shell where the various subshells and the possibility of Coster-Kronig transitions complicate the situation. In addition, there are no recent experimental values for krypton that would provide a check on theoretical values. Thus, a considerable uncertainty is introduced into the experimental ionization cross sections for the krypton L-shell.

EXPERIMENTAL APPARATUS AND METHODS

A general description of the experimental apparatus and methods is given in Appendix I. A more complete description is given below.

Apparatus

A schematic diagram of the Tandem Van de Graaff accelerator used to produce proton beams and the beam line on which the experiment was carried out is shown in Plate IV. H^- ions produced in the dc discharge source are magnetically selected and accelerated to the high voltage terminal. There, they are stripped of electrons and then accelerated away from the terminal to be magnetically analyzed by the 90° bending magnet. The magnet, which is calibrated by a nuclear magnetic resonance, provides a very narrow energy window for the beam, the spread in beam energy being less than 10 keV. The absolute calibration of the beam energy is determined from threshold nuclear reactions.

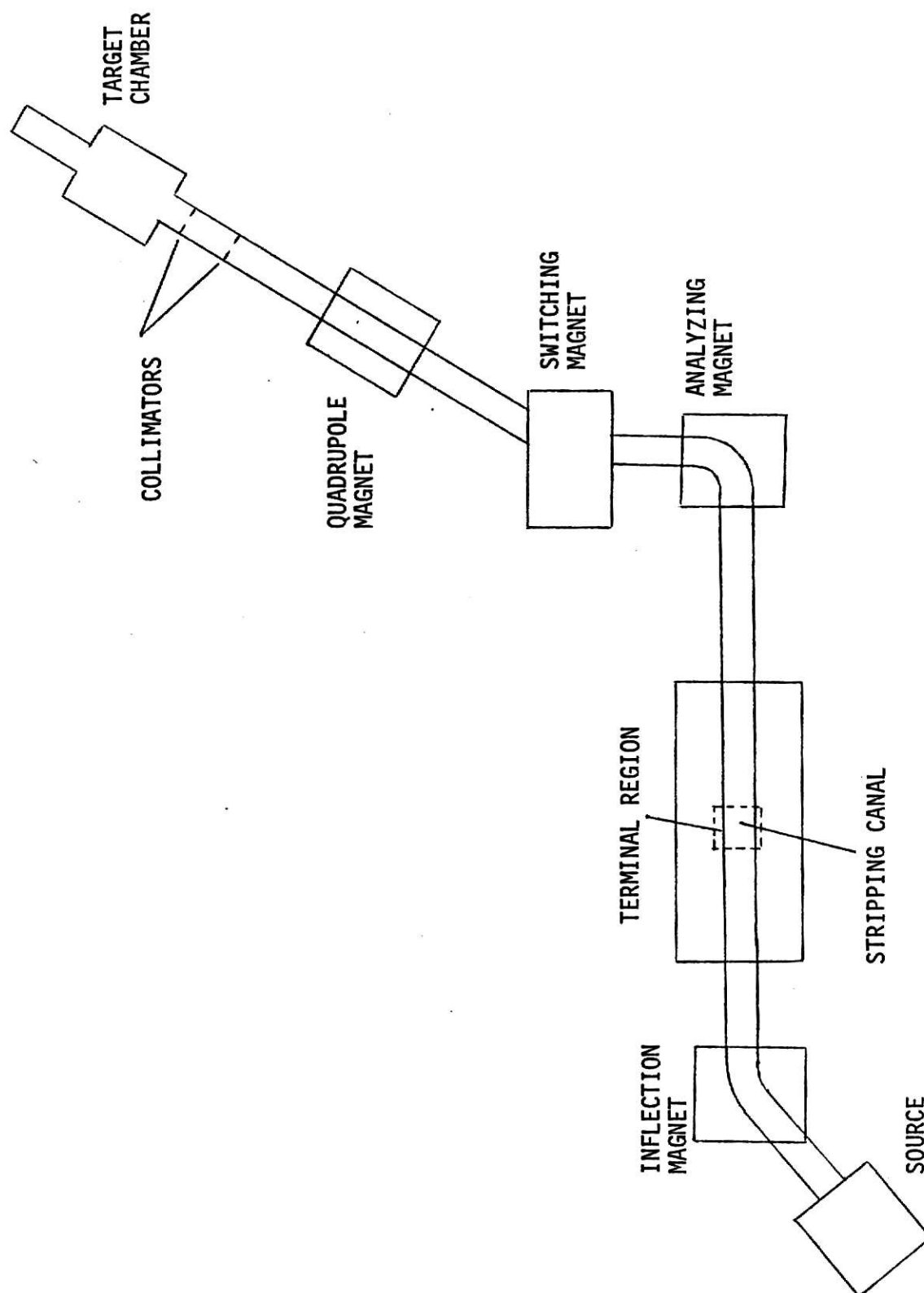
The beam is directed into the beam line by means of a switching magnet and is collimated by two sets of tantalum slits before reaching the target chamber. The target chamber, which houses the gas cell, is illustrated in Plate V. The gas cell is differentially pumped by means of two six-inch NRC diffusion pumps using DC-704 oil and liquid nitrogen traps. Foreline vacuums are maintained by two Welch 1376 mechanical roughing pumps. The residual pressure of the beam tube was typically 10^{-7} to 10^{-6} torr and was measured by an NRC 831 Vacuum Ionization Gauge.

Target gas pressures were regulated by means of an MKS Baratron Type 90 Capacitance Manometer coupled to a Granville-Phillips 213 Automatic Pressure Controller. In addition to being calibrated at the factory, the manometer has been calibrated in experimental use by a GM-100A McLeod gauge

EXPLANATION OF PLATE IV

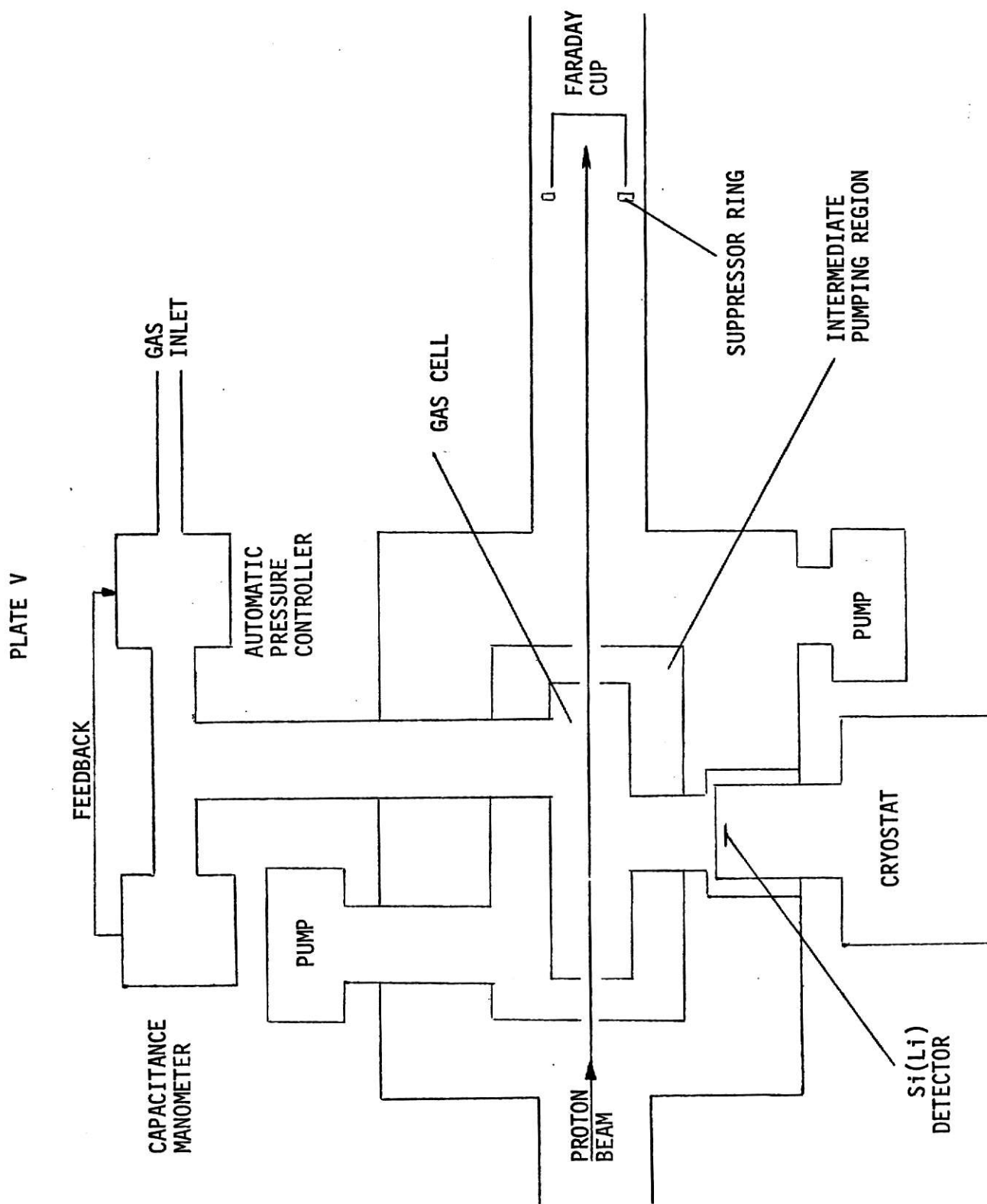
Schematic diagram of the Tandem Van de Graaff accelerator and the beam line on which the experiment was carried out.

PLATE IV



EXPLANATION OF PLATE V

Schematic diagram of the apparatus used to observe x-ray production by proton bombardment of gases.



supplied by Consolidated Vacuum Corporation and itself calibrated at the National Bureau of Standards. The accuracy of the manometer calibration is better than 10% at pressures between one micron and one torr.

The proton beam enters the gas cell through a pair of circular graphite apertures and exits the cell through a second pair of apertures larger in diameter than the first pair. This allows for a well-collimated beam and total beam transmission through the gas cell. The entrance apertures were chosen large enough to allow beam currents of 500 na to one μ a on target as well as to minimize slit-edge scattering. For the data presented in this paper, the aperture sizes from front-to-rear were 1.5 mm, 2.0 mm, 2.0 mm, and 3.0 mm. The beam was collected by a Faraday cup having a suppressor ring maintained at -300 V. Beam current was monitored and integrated by a Brookhaven 1000 current integrator.

X-rays resulting from collisions in the gas cell were detected by means of a Kevex 3000 liquid nitrogen cooled Si(Li) detector mounted inside the gas cell and at right angles to the beam direction. The detector, which was drifted to a depth of 3 mm, had an active area of 80 mm² and was maintained under separate vacuum by means of a .025 mm beryllium window. A bias voltage of -300 V was applied across the detector in normal operation. The amplification system for detector pulses consisted of a pulsed optical feedback preamplifier and a Kevex 4500 x-ray amplifier. Pulses from the amplifier were converted in a Geoscience Nuclear ADC and analyzed in a TMC 4096-channel analyzer. The data was stored on magnetic tape and transferred to a PDP-15 computer for analysis.

The resolution of the detector was typically 200 eV FWHM at 5.9 keV; however, this was highly dependent on noise vibrations picked up by the microphonic detector. This sensitivity to noise was reduced considerably

by shielding the liquid nitrogen dewar of the detector with foam rubber. Nevertheless, the close proximity of the Granville-Phillips pressure controlling valve to the detector still degraded the resolution by almost a factor of two when the valve was controlling. This is illustrated in Plate VI for the argon K x-ray peak. Because of this effect, it was necessary to turn off the pressure controller while data was accumulating. The resulting pressure drift could be kept small by carefully setting the valve and monitoring the output of the capacitance manometer continuously. A precision in the pressure setting of better than 2% was maintained for all runs.

A removable iron-55 source mounted within the gas cell was used to energy-calibrate the detector. This source gave two well-separated peaks: the Mn $K\alpha$ at 5.895 keV and the Mn $K\beta$ at 6.492 keV. A pulse height spectrum of these peaks is shown in Plate VII. In addition, sample spectra of the Ar K, Kr K, and Kr L lines are shown in Plates VIII, IX, and X respectively. Using the midpoints of the Mn K peaks as well as that of the proton-induced $K\alpha$ line of argon, a calibration curve was obtained by using a linear least squares fitting routine. A graph of this curve is shown in Plate XI. The curve was used to determine the experimental energies of the lines listed in Table II. Comparison was made with the weighted energies given by Storm and Israel⁴⁰. Agreement is, in general, excellent with the greatest deviations occurring for the $K\alpha$ and $K\beta$ lines of krypton and amounting to less than 20% of a peak width.

Five background x-ray peaks were also observed, and the above calibration was used to identify them as Cr $K\alpha$ (5.411 keV), Cr $K\beta$ (5.947 keV), Fe $K\alpha$ (6.400 keV), Fe $K\beta$ (7.059 keV), and Ni $K\alpha$ (7.472 keV). These were believed to originate from Coulomb excitation and fluorescence excitation of

TABLE II

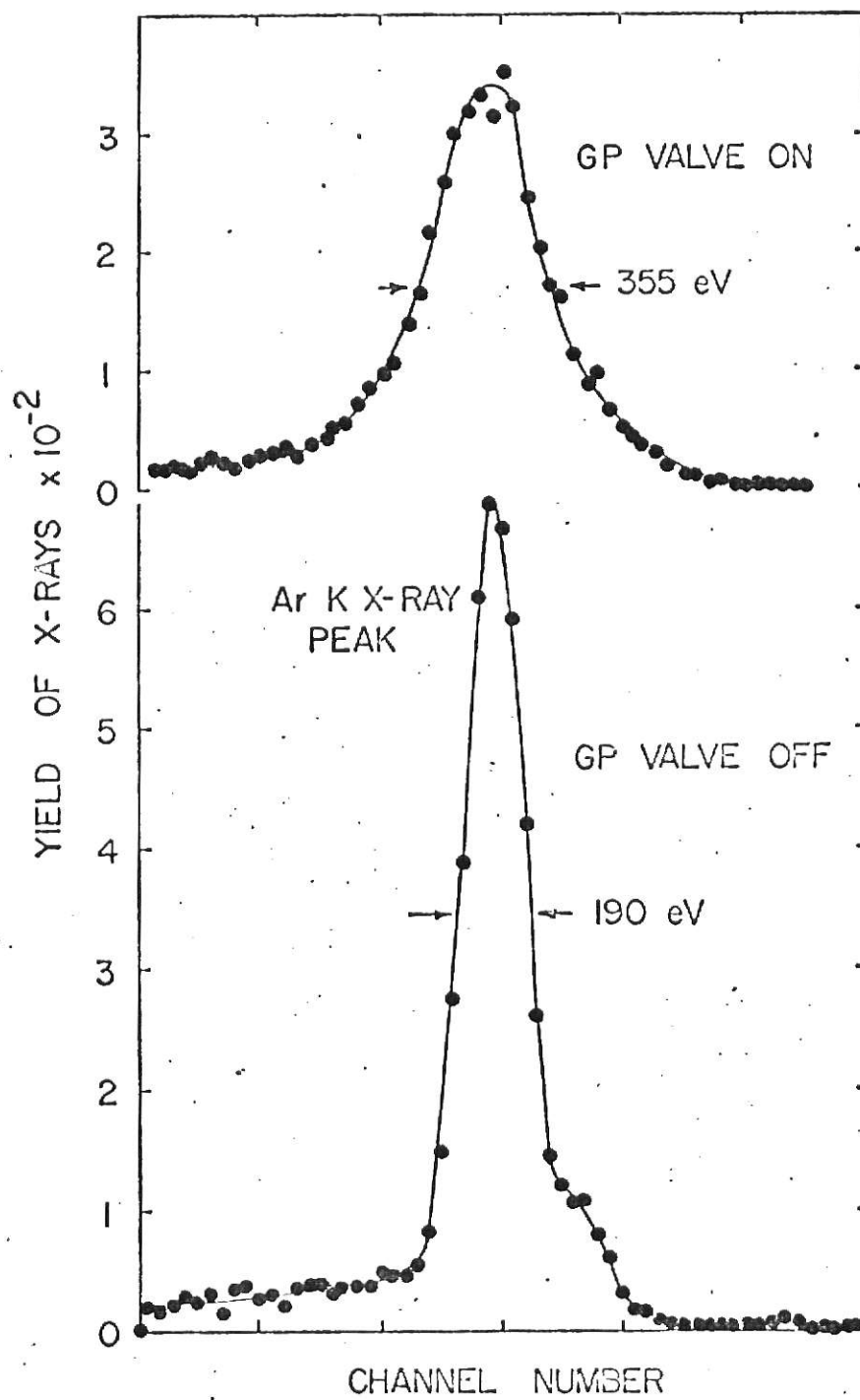
Comparison of Experimental and Weighted X-ray Energies

X-ray Line	Experimental X-ray Energy, E_x (keV)	Weighted X-ray Energy, E_w^{40} (keV)	$E_x - E_w$ (keV)
Kr L	1.616	1.631	-.015
Ar $K\alpha$	2.957	2.957	.000
Ar $K\beta$	3.179	3.191	-.012
Mn $K\alpha$	5.896	5.895	.001
Mn $K\beta$	6.491	6.492	-.001
Kr $K\alpha$	12.675	12.630	.045
Kr $K\beta$	14.190	14.126	.064
Experimental calibration: E_x (keV) = .0394 x (Channel number) - .1702			

EXPLANATION OF PLATE VI

A comparison of resolution on the argon K x-ray peak with the Granville-Phillips valve controlling (upper spectrum) and with the valve not controlling (lower spectrum). The incident proton energy is 3.0 MeV, and the manometer pressure is one micron.

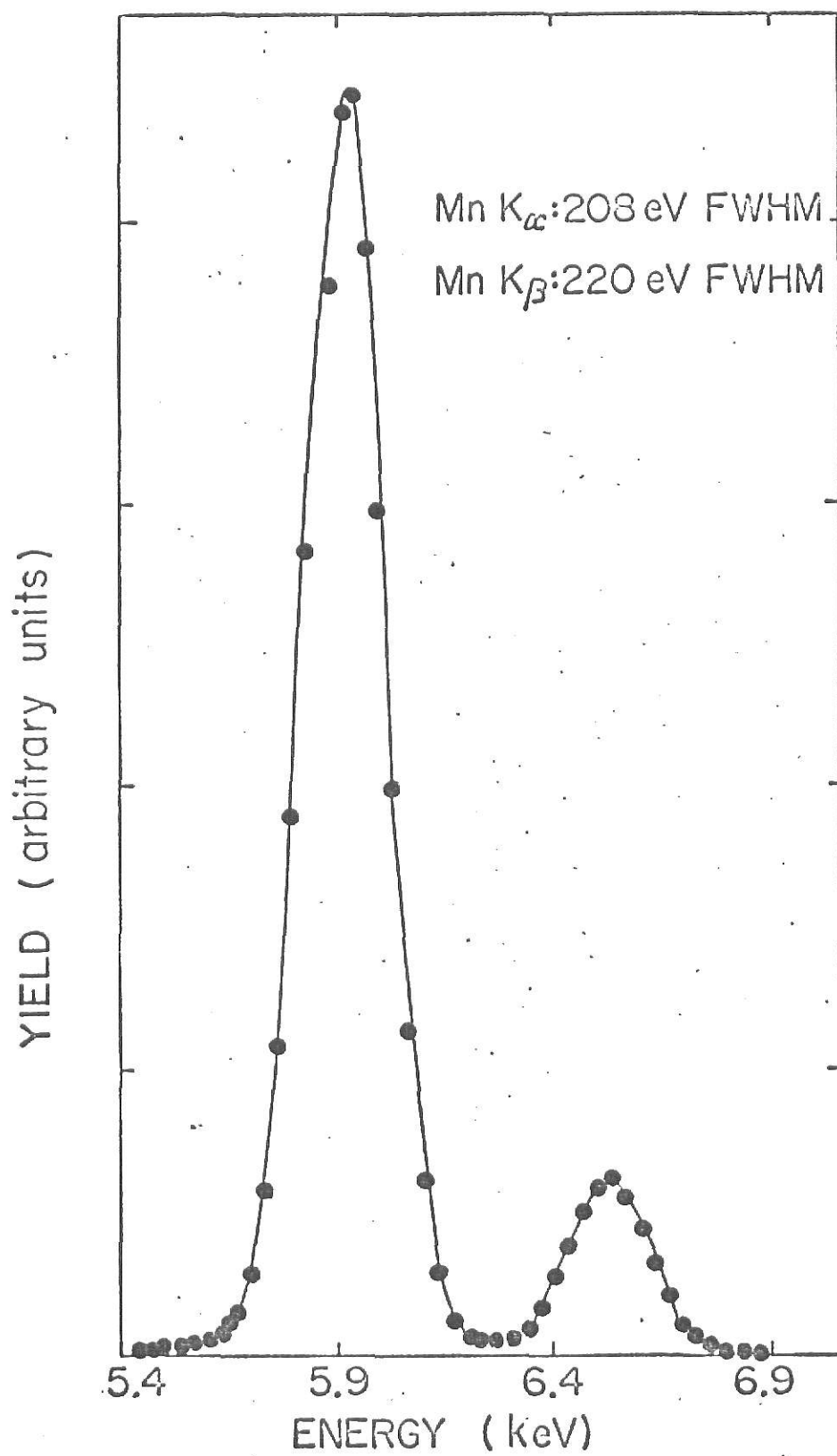
PLATE VI



EXPLANATION OF PLATE VII

Pulse height spectrum of the iron-55 source. The large peak is the Mn $K\alpha$ at 5.895 keV, and the small peak is the Mn $K\beta$ at 6.492 keV.

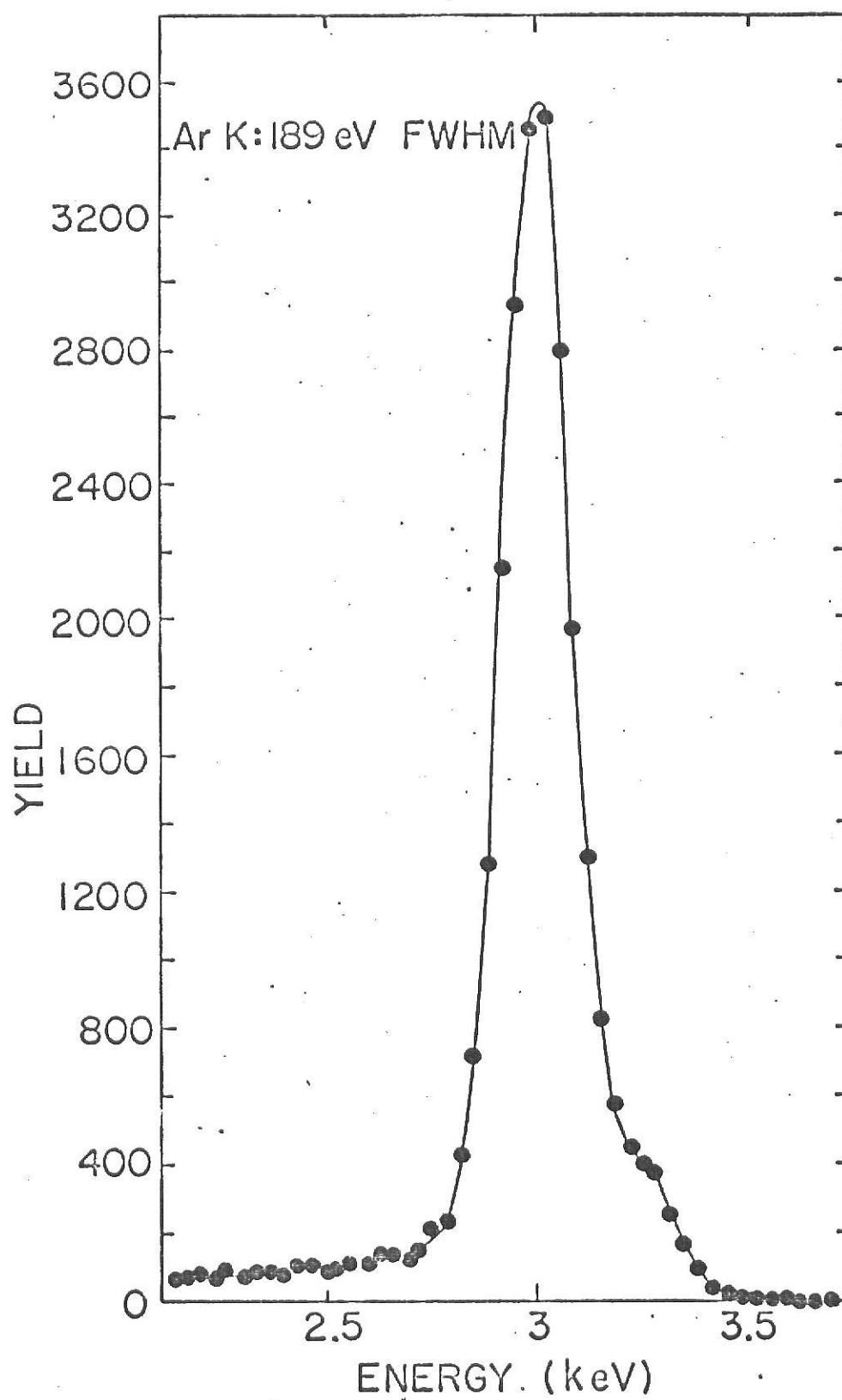
PLATE VII



EXPLANATION OF PLATE VIII

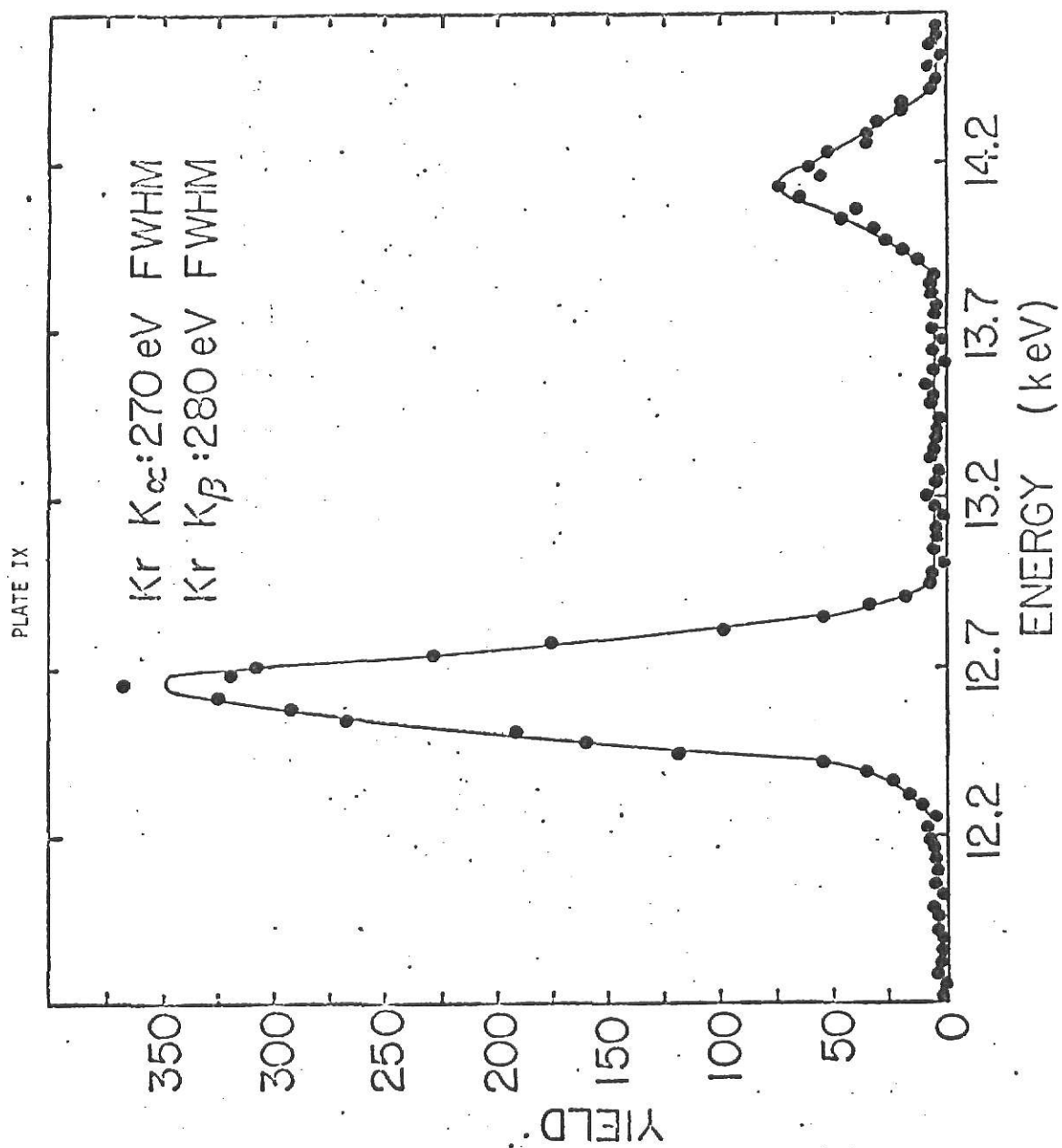
Pulse height spectrum of the argon K x-ray lines. The main peak is the $K\alpha$ at 2.957 keV, and the shoulder on the high energy side of the $K\alpha$ line is the $K\beta$ line at 3.191 keV.

PLATE VIII



EXPLANATION OF PLATE IX

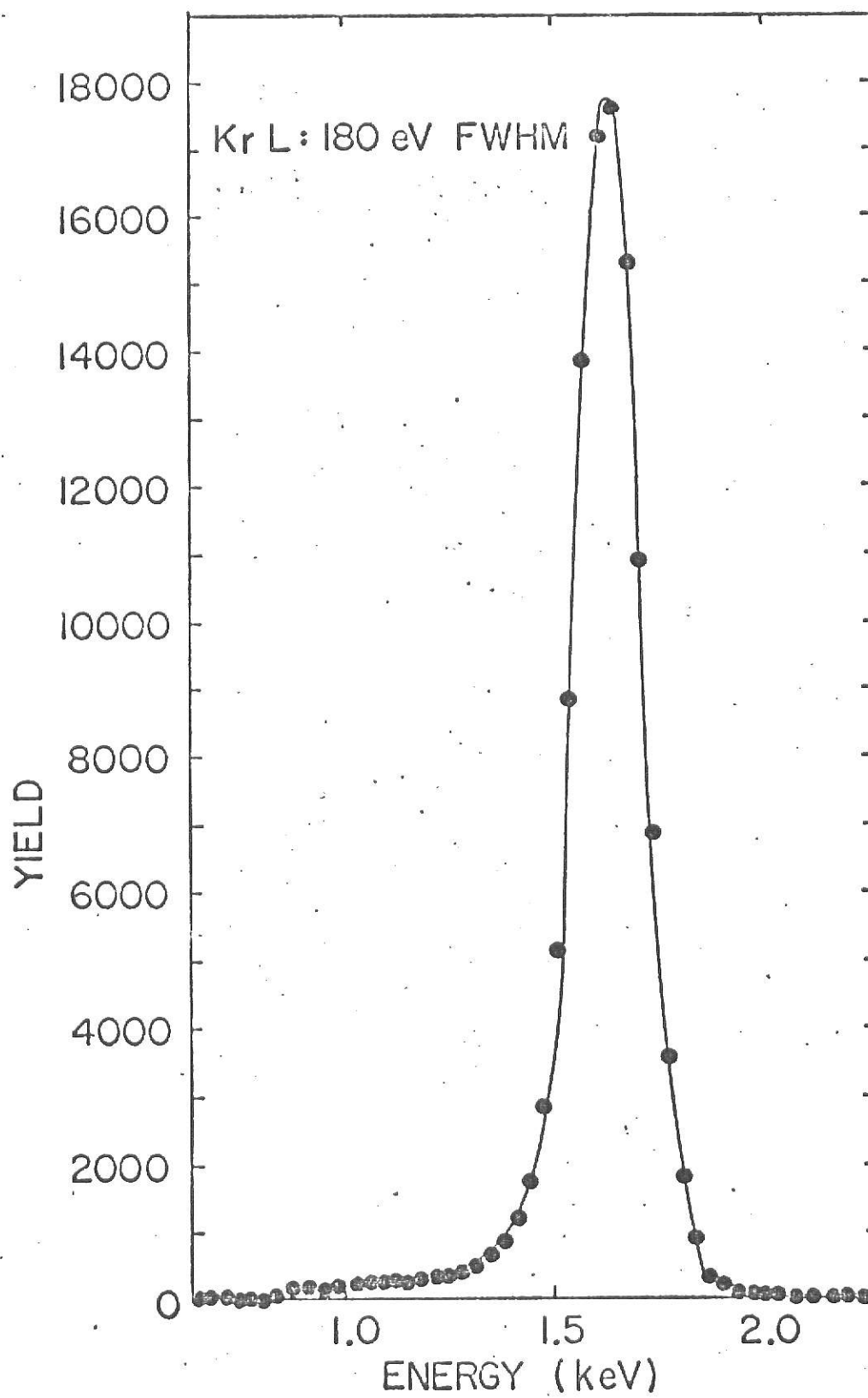
Pulse height spectrum of the $K\alpha$ and $K\beta$ lines of krypton. The larger peak is the $K\alpha$ at 12.63 keV, and the smaller peak is the $K\beta$ at 14.126 keV.



EXPLANATION OF PLATE X

Pulse height spectrum of the krypton L x-rays. The energy of the peak is 1.631 keV.

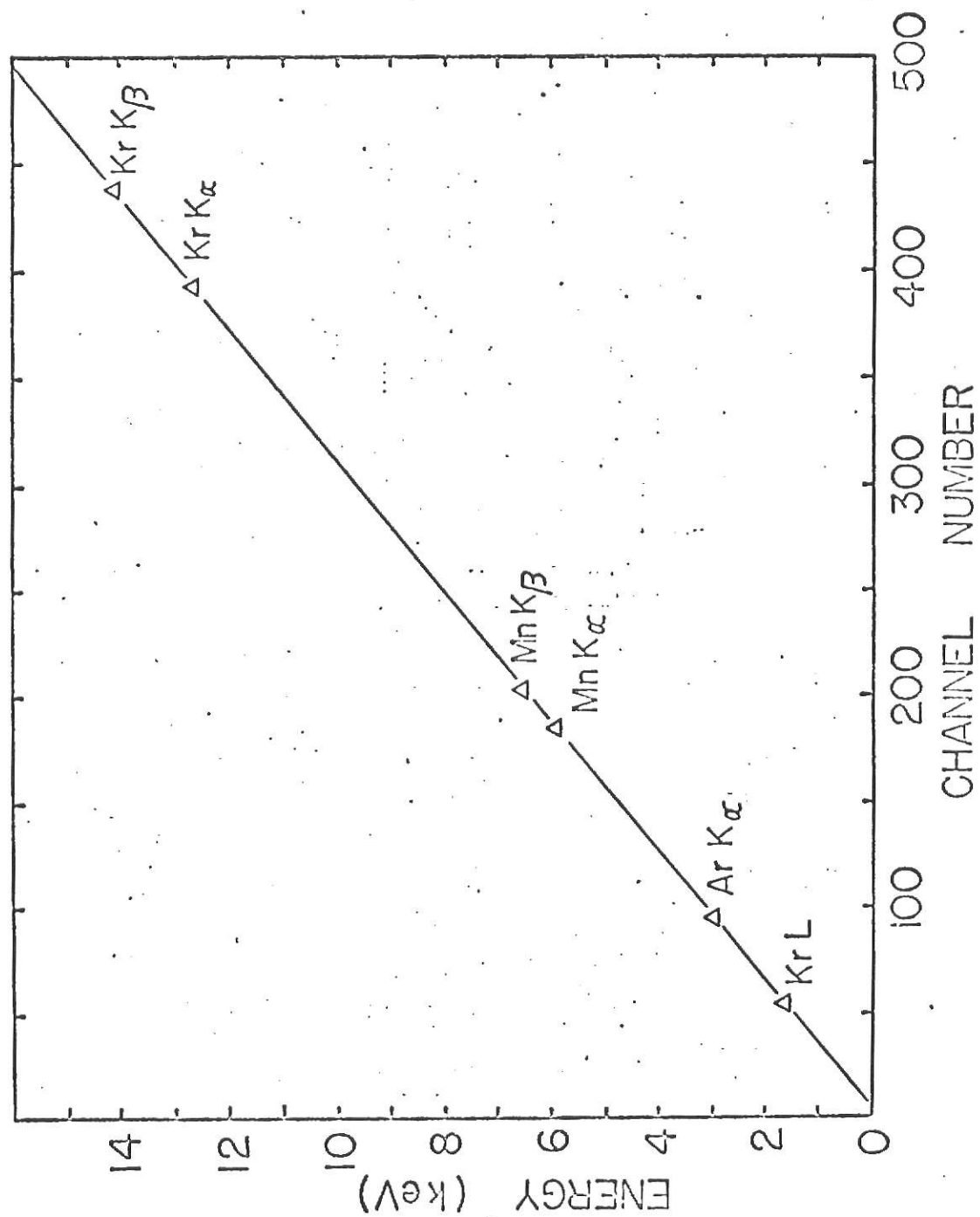
PLATE X



EXPLANATION OF PLATE XI

Calibration curve for identification of x-ray lines. The curve was determined from weighted x-ray energies given by Storm and Israel⁴⁰ for the Mn K α , Mn K β , and Ar K α lines.

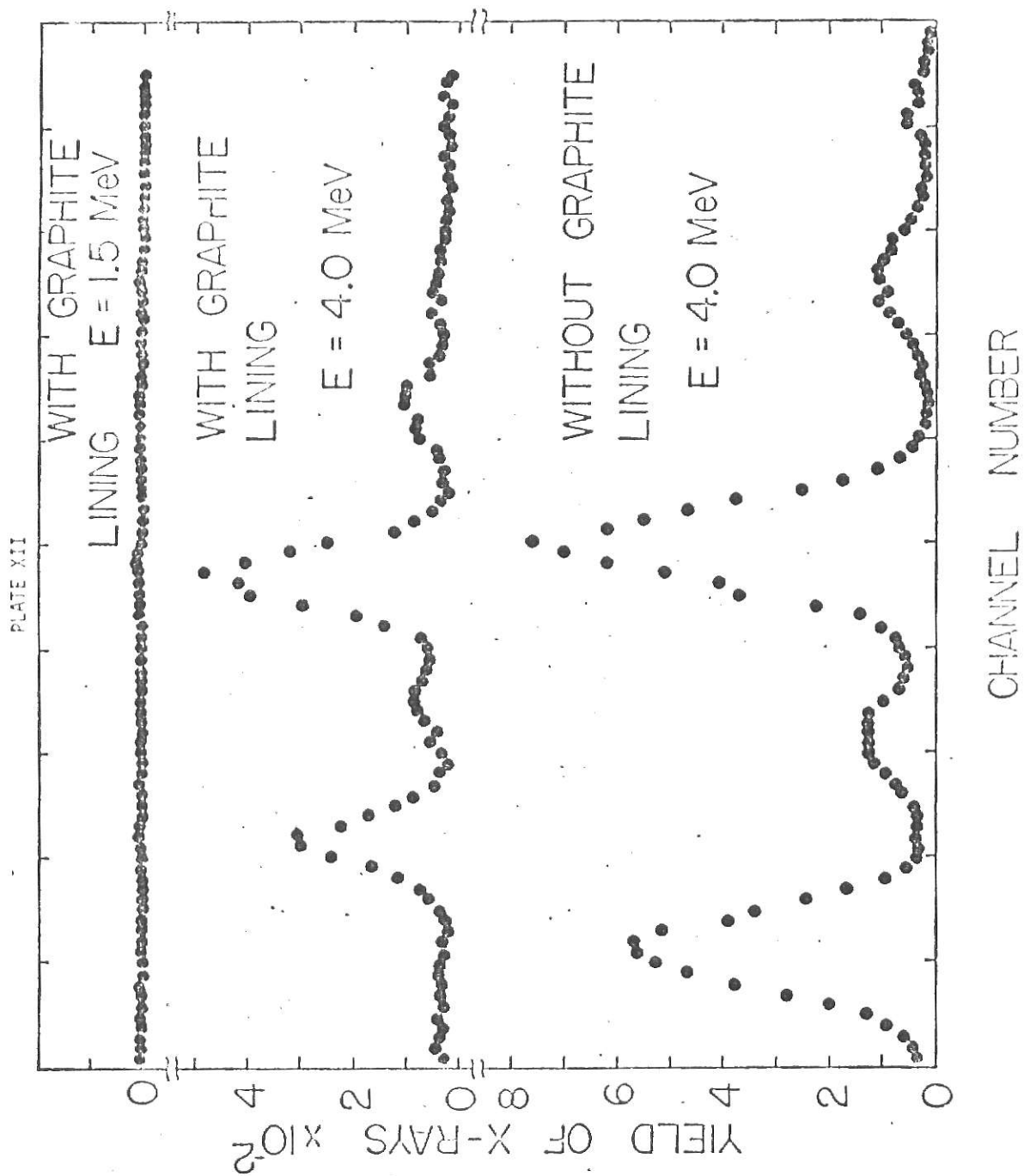
PLATE XI



the stainless steel from which the gas cell was made. This background was reduced by lining the gas cell with graphite, which emits x-rays too low in energy to be seen by the detector. From Plate XII, it can be seen that at low incident proton energies, the background peaks are quite small; however, for the highest incident energies, the peaks are large even with the graphite lining. In addition to the characteristic x-rays, a continuum background of x-rays was observed. Plates XIII and XIV illustrate that this continuous background is significantly greater for high incident energy than for low. Plate XIII makes this comparison for the background in the vicinity of the Kr L x-ray peak for the same counting rate conditions in the peak and incident energies of 1.5 and 4.5 MeV; whereas, Plate XIV shows background spectra for an evacuated gas cell and incident energies of 1.5 and 4.5 MeV. This increase of background with incident energy is attributed to scattering of high energy gamma rays from nuclear reactions in the various collimating slits. In particular, with graphite apertures, the background becomes prohibitively large when the proton energy is above 4.8 MeV, in which case the inelastic scattering channel contributes to the background. The apertures were originally of tantalum, but this resulted in a large background even at low energy and they were replaced by the graphite ones. In order to continue this experiment at energies above 5 MeV, it will be necessary to insure that the beam is collimated before the gas cell and that the slits intercepting the beam are thoroughly shielded from the detector.

EXPLANATION OF PLATE XII

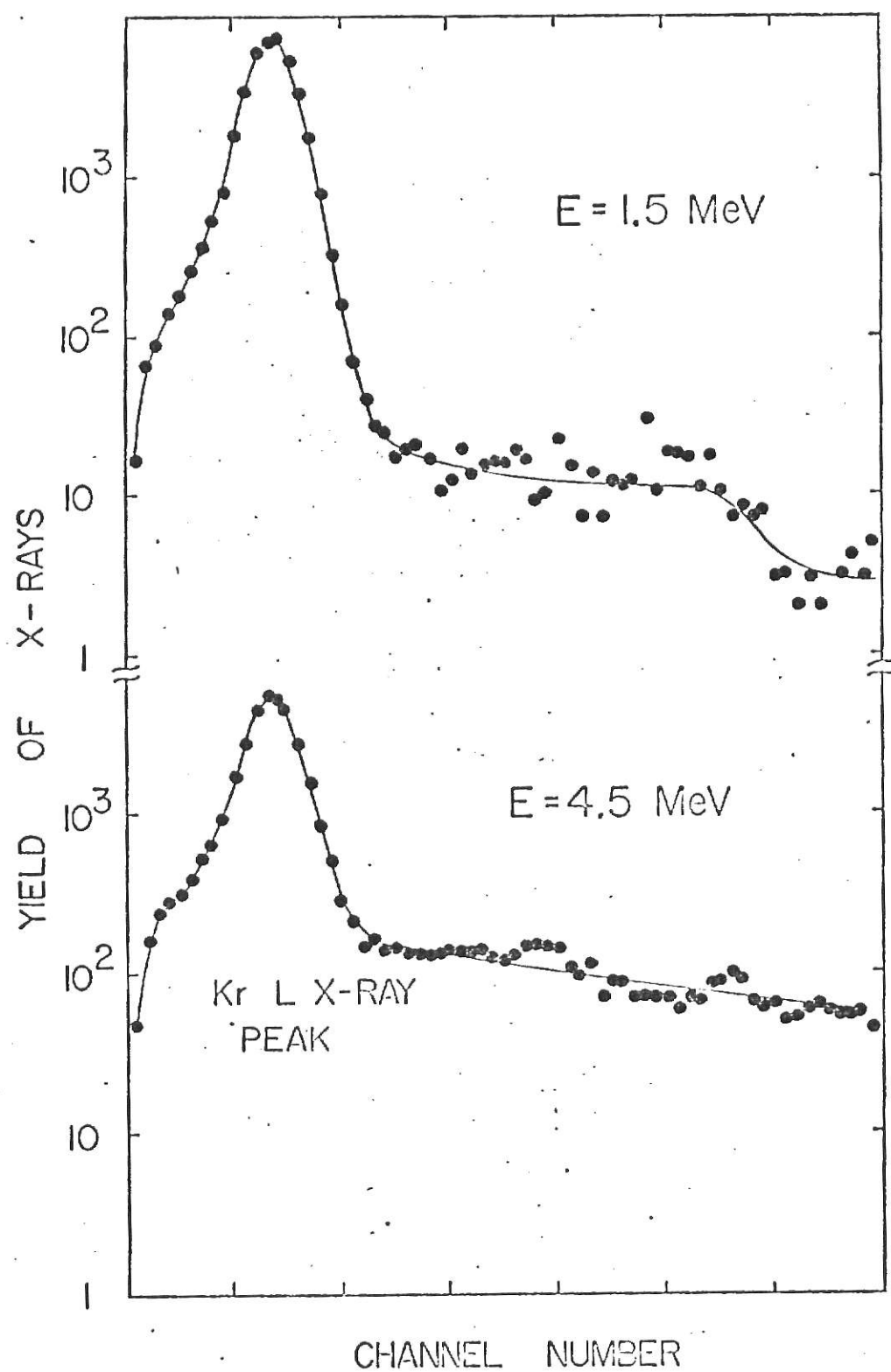
Background peaks resulting from excitation of the stainless steel gas cell. The upper spectrum is taken with graphite lining the gas cell at an incident energy of 1.5 MeV, the middle spectrum with graphite lining at an incident energy of 4.0 MeV, and the lower spectrum without graphite lining at an incident energy of 4.0 MeV. All spectra were taken for the same amount of collected beam. (Note that the energy calibration for the upper two spectra is different than that for the lower spectrum.)



EXPLANATION OF PLATE XIII

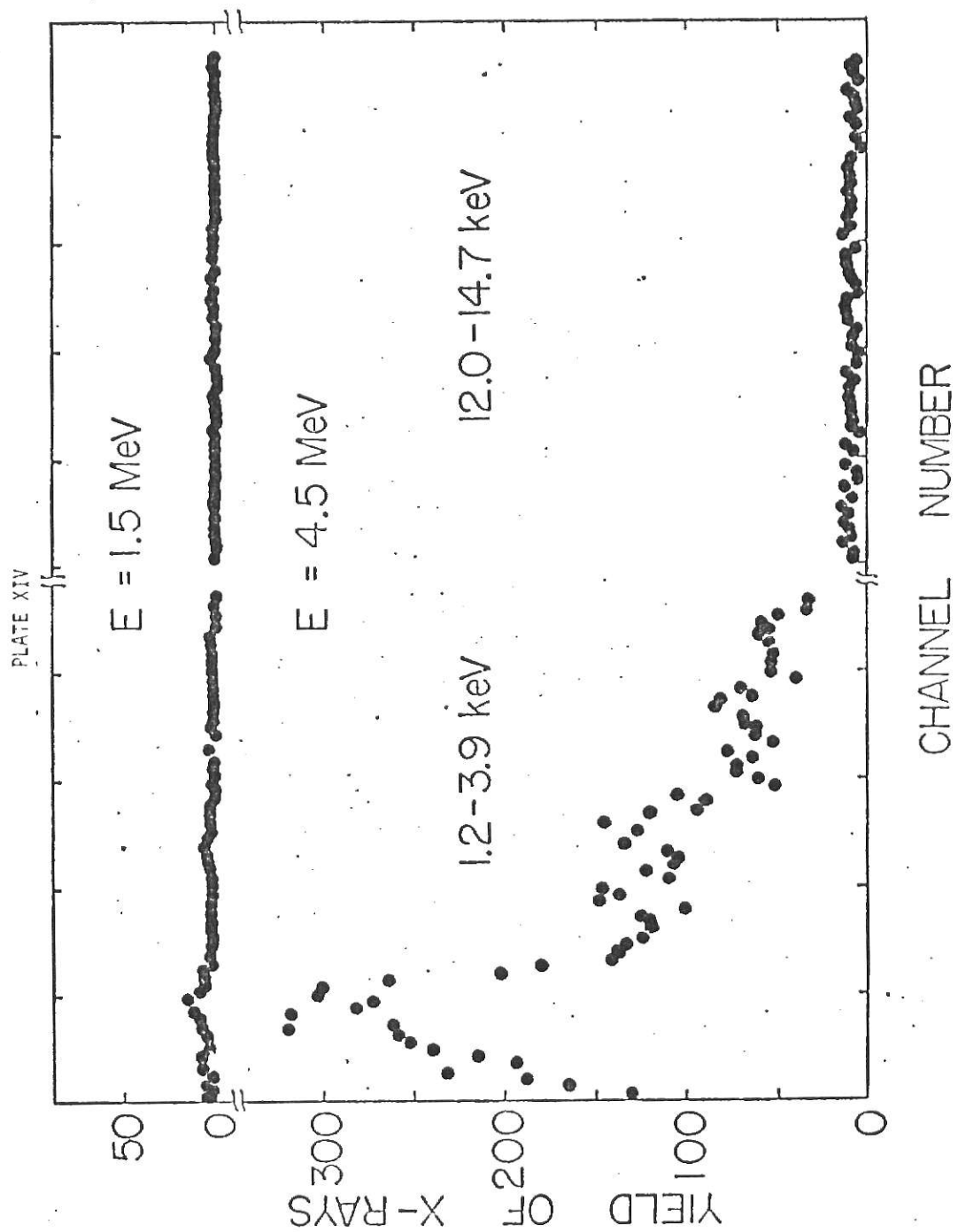
Comparison of the continuous background in the vicinity of the krypton L x-ray peak for incident proton energies of 1.5 and 4.5 MeV. Counting rate conditions in the peak are the same for both spectra.

PLATE XIII



EXPLANATION OF PLATE XIV

Comparison of the continuous background at residual gas cell pressures for incident proton energies of 1.5 and 4.5 MeV. The low energy and high energy portions of the x-ray spectra are shown. The amount of beam collected is the same for both spectra.



Corrections Applied to X-ray Yields

Single Collisions

The extraction of x-ray production cross sections from thick target yields is complicated by the multiple collisions which an incident proton may undergo. To correct for this, one must utilize stopping powers as well as the slope of the excitation function. However, if the target is thin enough so that single collision conditions prevail, then these corrections are unnecessary since each incident proton produces one x-ray at most and the energy of the proton is essentially unchanged in the gas target. These conditions were maintained in the present experiment by operating in a range of target pressures for which the growth of x-ray yield with pressure was linear. For gas pressures less than five microns--the region in which x-ray yields were normally obtained--this criterion was indeed satisfied. In fact, thin target conditions were found to prevail up to manometer pressures of at least 30 microns, as illustrated in Plate XV for krypton L x-ray yields. Thus, no correction to x-ray yields for multiple collision effects was required.

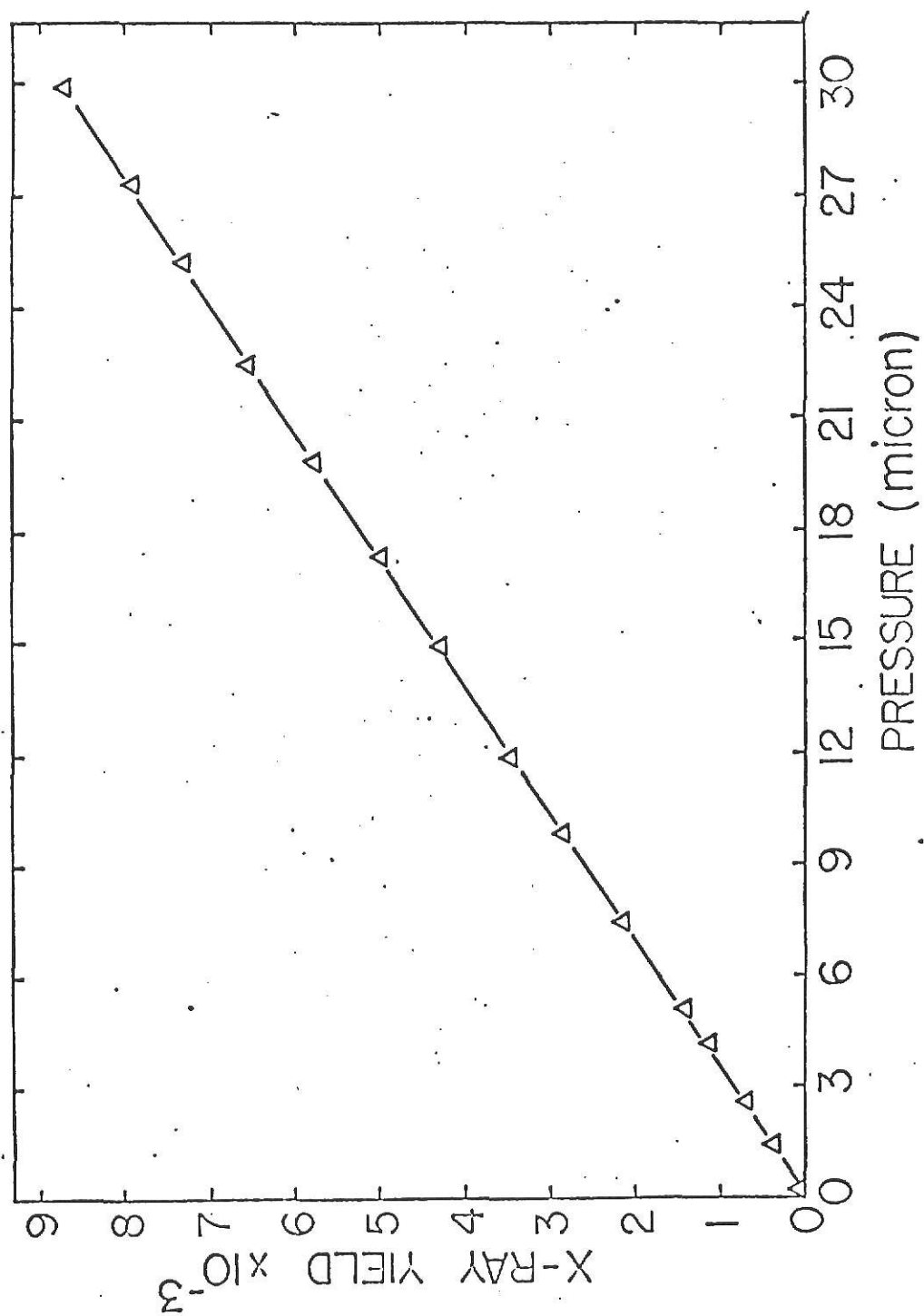
Electronic Dead Time

The use of the pulsed optical feedback system made necessary a significant dead-time correction to x-ray yields. This correction was achieved electronically by use of the circuit shown in Plate XVI. The amplifier gating pulse restricted the counting of integrated beam current to detector live time only. Inversion of the gating pulse also made it possible to count for dead time only, and a comparison of this dead time result with the total time provided a check on the live time result. At

EXPLANATION OF PLATE XV

Yield of krypton L x-rays as a function of manometer pressure. The yields have been normalized to one microcoulomb collected beam and corrected for electronic dead time. The incident proton energy is 3 MeV.

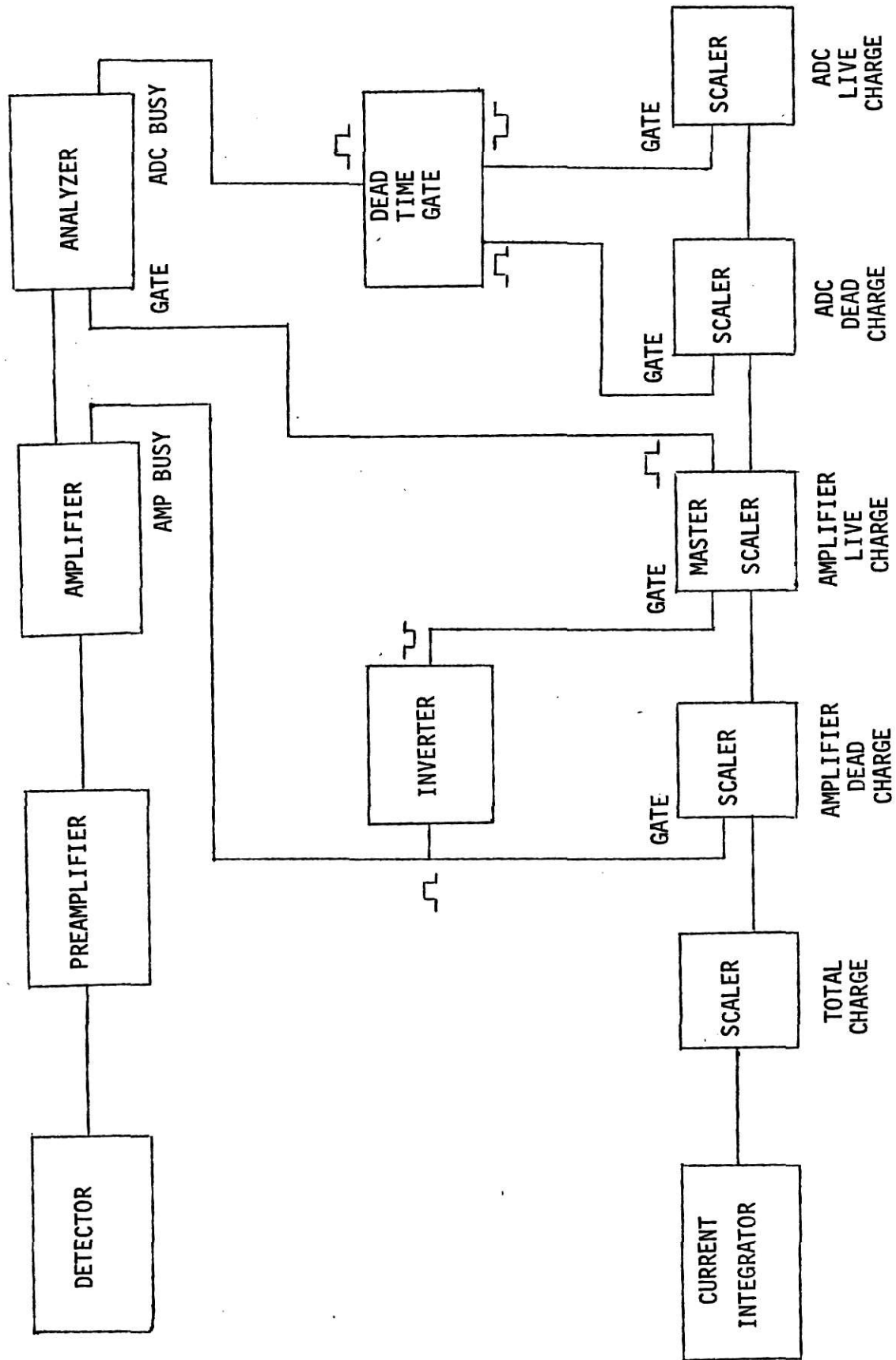
PLATE XV



EXPLANATION OF PLATE XVI

Electrical circuit used to correct the x-ray yields for amplifier dead time as well as ADC dead time.

PLATE XVI



the highest counting rates, corrections of up to 40% were applied by using this method. The accuracy of this correction technique was tested by using two different amplification systems to observe the Mn K x-rays from the iron-55 source. One of the systems was that described above while in the other system, the Kevex amplifier was replaced by an Ortec 450 amplifier which was not gated off during feedback and needed no dead time correction. The x-ray yields obtained at high counting rates agreed to within 2% for the two systems.

A dead time correction technique similar to that used for the Kevex amplifier was also used for the ADC. This is shown in Plate XVI. The ADC dead time correction was typically less than 1% except at the highest counting rates for which it was less than 3%.

Target and Detector Geometry

If one assumes that x-ray emission from the target is isotropic, then it becomes a relatively simple matter to correct the x-ray yields for the solid angle subtended by the detector. The validity of this assumption has been shown to 2% by Bernstein and Lewis⁴¹ for emission of L x-rays of gold, and it is assumed to be valid in all cases.

If the total interaction length, L' , seen by the detector is small compared to the distance, x , from beam to detector, then a good approximation to the solid angle is

$$\Omega = L' \Omega_0 \quad (2.1)$$

where $\Omega_0 = A/4\pi x^2$, A being the active area of the detector. It is assumed that the interaction region is a line source. For our geometry, the detector was quite close to the beam so that a better approximation

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was deemed necessary.

A diagram of the geometry is shown in Plate XVII with the relevant distances labeled. L is the interaction length seen by all points of the detector, and $2l$ is the interaction length seen by fractional portions of the detector so that $L' = (L + 2l)$. It can easily be proven that

$$L = (a - d_0 r)/(1 - r)$$

$$\text{and } l = d_0 r/(1-r)$$

where $r = \Delta/x$. Now each point on the length L is the vertex of a cone of solid angle subtended by the detector. Integration of this cone over L will then give the contribution of L to the total solid angle correction. The result to first order is

$$\Omega' = L\Omega_0 \cos \alpha_0$$

where $\cos \alpha_0 = [1 + (L/2x)^2]^{-1/2}$. If $(L/2x)^2$ is small, a good approximation to this result is

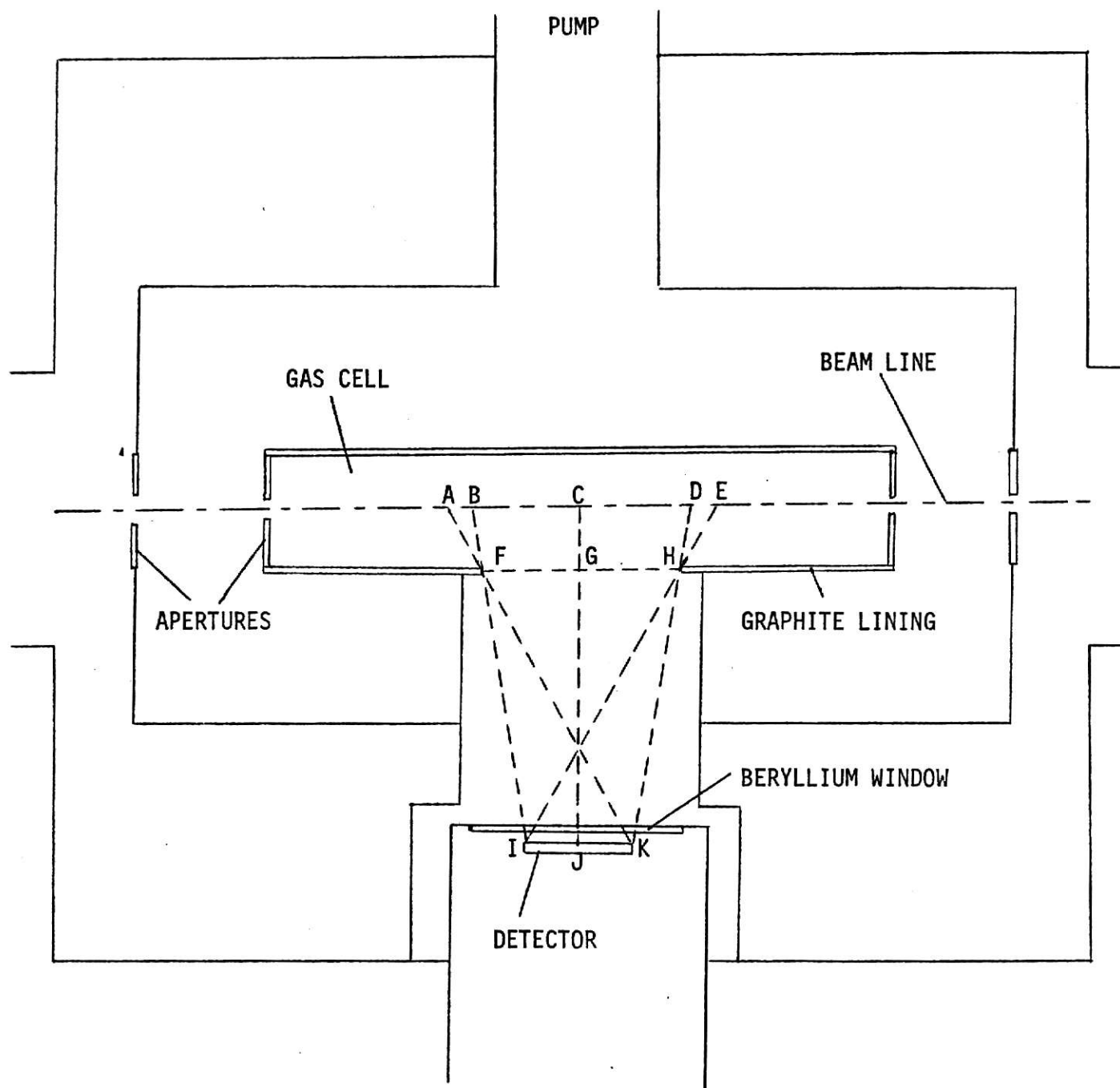
$$\Omega' = L\Omega_0 (1 - L^2/8x^2) \quad (2.2)$$

where $\cos \alpha_0$ has been expanded in a binomial series. For the geometry of this experiment, the first-order term of (2.2) results in a contribution of about 0.8% to Ω' . A somewhat larger contribution, however, comes from the length $2l$. By following a similar procedure as for L but taking into account the fact that the detector area seen is different for each point on l , one obtains $l\Omega_0$ as the contribution--in first approximation--to the solid angle correction due to $2l$. This contribution amounts to about 5% of the total solid angle. For our geometry, then, an excellent approximation to the total solid angle correction used to determine x-ray yields from the gas cell is

EXPLANATION OF PLATE XVII

Target and detector geometry for which solid angle corrections were determined.

PLATE XVII



$$\overline{AB} = \overline{DE} = 1$$

$$\overline{BD} = L$$

$$\overline{FH} = a$$

$$\overline{CG} = \Delta$$

$$\overline{CJ} = x$$

$$\overline{IK} = d_0$$

$$\Omega_t = \Omega_o(L + 1 - L^3/8x^2). \quad (2.3)$$

Using $A = .80 \text{ cm}^2 \pm 1\%$, $x = 7.66 \text{ cm} \pm 5\%$, $L = 1.54 \text{ cm} \pm 1.7\%$, and $l = .08 \text{ cm} \pm 9.4\%$, one obtains for this experimental geometry $\Omega_t = .00175 \text{ cm} \pm 7\%$. It should be noted that the above development assumes a well-defined position for the detector. If, however, the detector is located in such a way that part of it is shadowed from the interaction region, then the above value of Ω_t will be too large by some amount. From the present knowledge of the detector geometry, however, there is no reason to believe that this is the case.

Target Thickness

The number density of gas atoms used to convert the x-ray yields to single-atom yields was $9.67 \times 10^{15} P_T / (293 + \Delta T) \text{ cm}^{-3}$ where P_T is the target gas pressure in microns and ΔT is the deviation of gas temperature from 200°C and is neglected.

P_T , however, is not the same as the pressure, P_M , indicated by the manometer because the flow of gas through the large gas cell apertures sets up a pressure gradient between the manometer and the cell. An approximate analysis of this effect provides a means of experimentally determining the magnitude of the necessary corrections to P_M .

Fig. 1 is a schematic diagram of the target chamber with the pertinent quantities labeled. S_M is the unknown pumping speed from manometer to cell, S_1 and S_2 are the pumping speeds through the inner apertures, and P_I is the pressure of the intermediate region. Conservation of mass requires that

$$(P_M - P_T)S_M = (P_T - P_I)(S_1 + S_2).$$

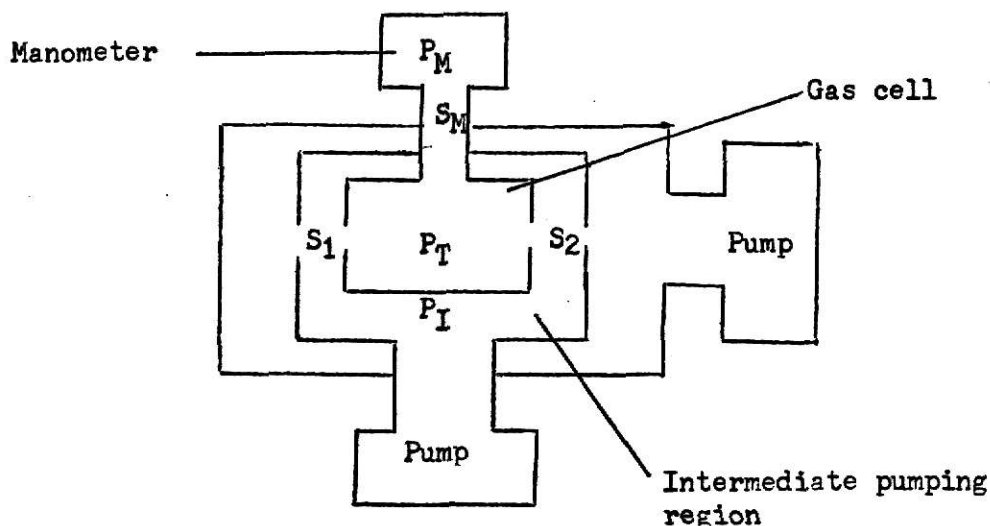


Fig. 1. Schematic diagram of the target chamber

Assuming that $P_I \ll P_T$ and setting $S_1 = S_2 = S$, one obtains an equation for P_M :

$$P_M = (1 + 2S/S_M)P_T.$$

Now the dependence on the particular target gas divides out in the ratio S/S_M . In addition, S is closely proportional to d^2 where d is the aperture diameter⁴². Then one obtains for P_T :

$$P_T = (1 + bd^2)^{-1}P_M \quad (2.4)$$

where b is a gas-independent constant.

The gas cell pressure, P_T , is proportional to I , the observed yield of x-rays. Under thin target conditions, one can experimentally determine the straight-line slope $Y = \Delta I / \Delta P_M$ for a given aperture diameter, d , and manometer pressure, P_M . Then Y is related to d according to

$$1/Y = c(1 + bd^2)$$

where c is a constant which depends on the x-ray production cross section. For five different aperture sizes ranging from $d = 1.0$ mm to 3.0 mm, the value of Y was measured for Ar K, Kr K, and Kr L x-rays. The incident proton energy was 3.0 MeV in all cases. For the three lines, the value of b was extracted using a linear least squares fitting routine. (It should be noted that the data for argon at $d = 1.0$ mm was not included in the fit, because the alignment conditions under which it was taken are believed to be different than those for the remaining data.) The results of the fitting procedure are plotted in Plate XVIII.

The values of b obtained were in reasonable agreement for the three lines. They were $.131 \pm 10\%$ for krypton L x-rays, $.118 \pm 15\%$ for krypton K x-rays, and $.138 \pm 15\%$ for argon K x-rays. The value $b = .13$, which is believed to be accurate to 15% , was chosen to make the pressure correction. For the data presented in the last section of this paper, the value of d was 2.0 mm so that a correction of $52 \pm 7\%$ was applied to P_M in order to obtain P_T .

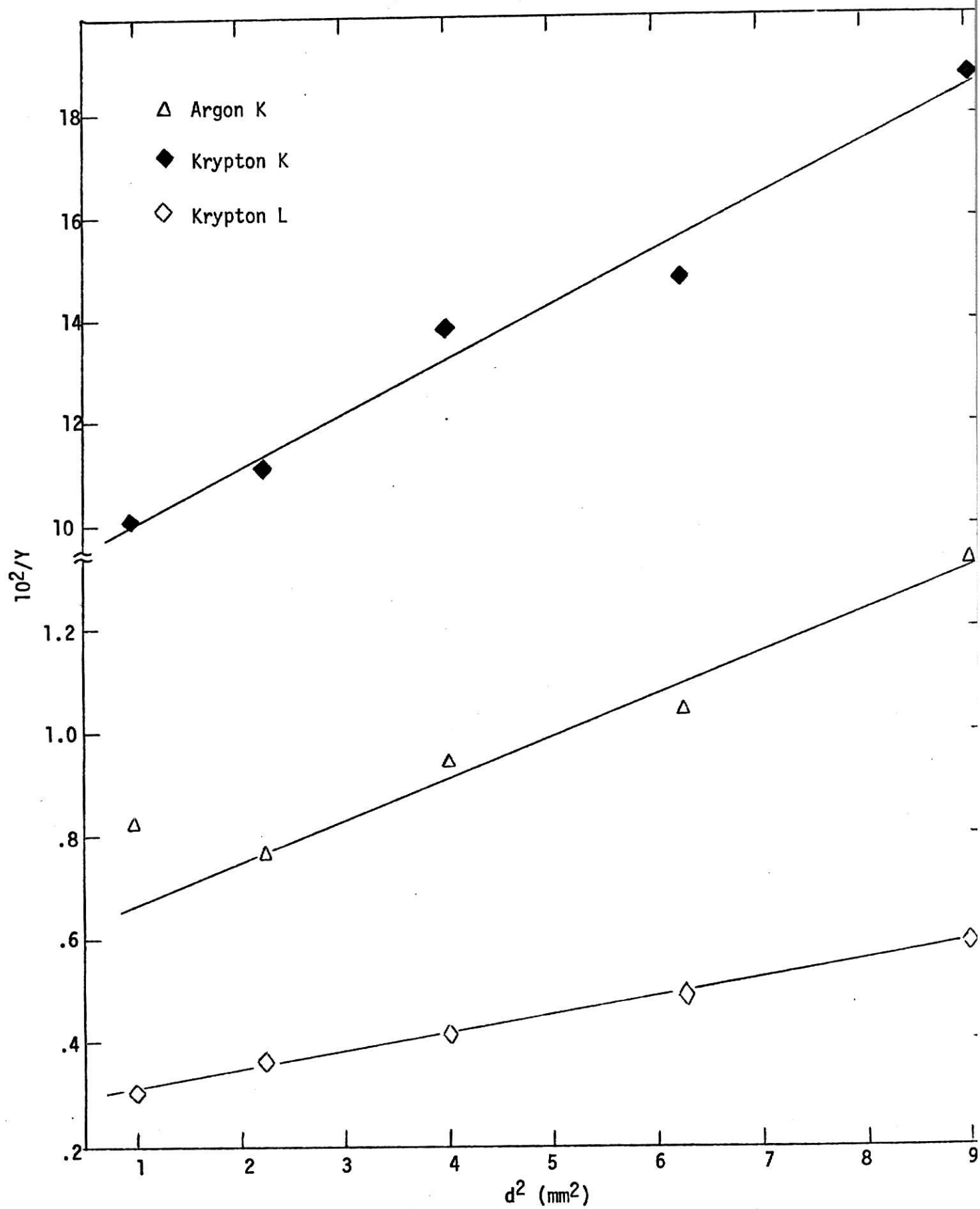
Absorption of X-rays and Detector Efficiency

For x-rays below 5 keV in energy, absorption between source and detector is an important factor in obtaining x-ray production cross sections. Absorption occurs in the target gas, the beryllium window, and the gold and insensitive silicon layers of the detector. Self-absorption in the target is negligible and amounts to less than $.01\%$ --as determined from mass absorption coefficients of Storm and Israel⁴⁰--for all the x-rays observed in this experiment. However, absorption in the remaining three

EXPLANATION OF PLATE XVIII

$10^2/Y$ versus the square of the gas cell aperture diameter, d , for argon K x-rays (triangles), krypton K x-rays (closed diamonds), and krypton L x-rays (open diamonds). Y is the change in the observed yield of x-rays with respect to manometer pressure.

PLATE XVIII



layers must be taken into account in determining detector efficiency.

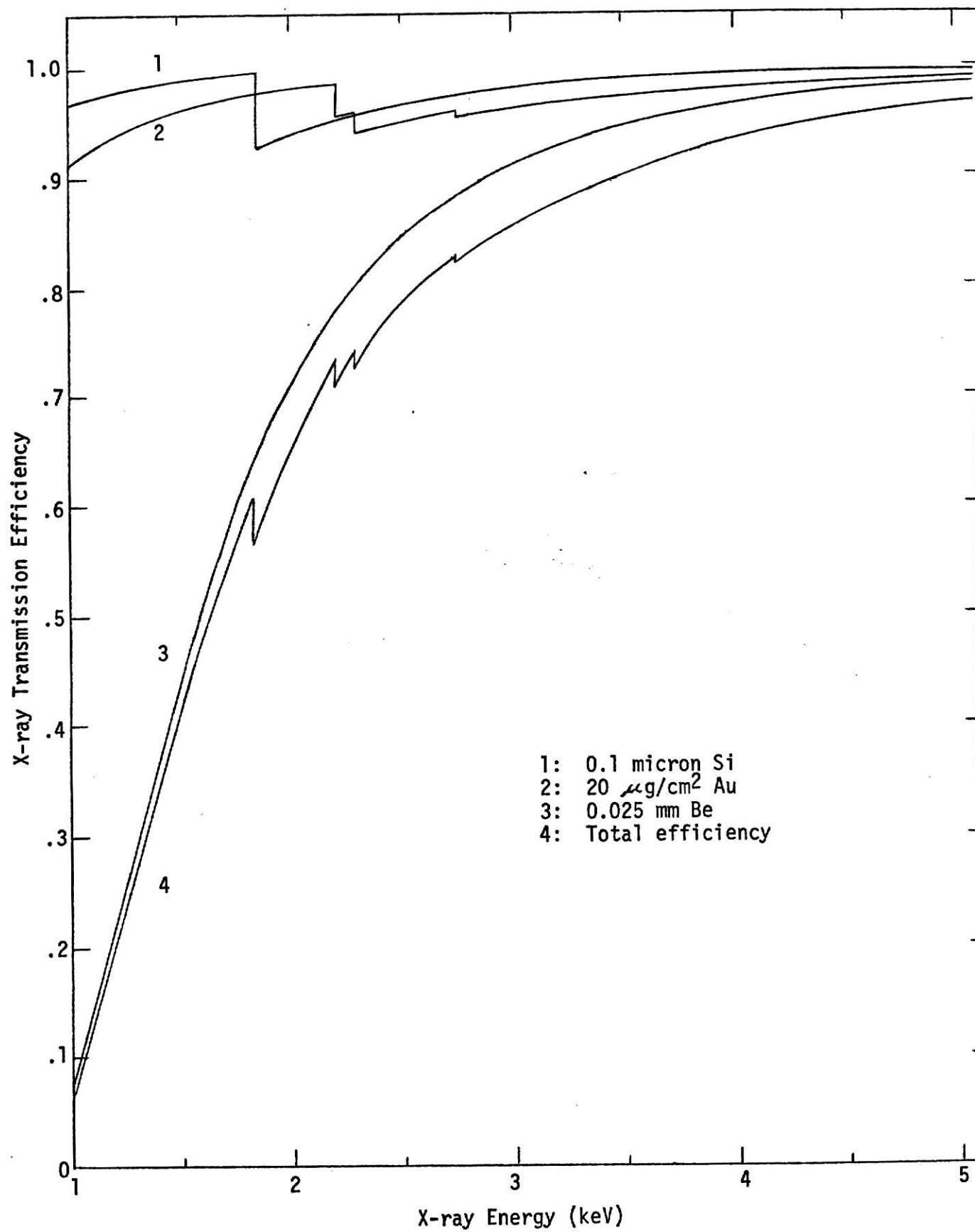
Published experimental efficiency curves for Si(Li) detectors have been obtained^{27,43}; however, these do not cover the energy region of interest. This lack of absolute efficiency standards is a fundamental problem which has not been solved for low energy x-ray detectors in general. The experimental uncertainties associated with the higher energy efficiency curves suggest that a theoretical calculation may be as accurate. The problem with this type of calculation is that the thicknesses of the absorbing layers, in particular the gold and silicon layers, are not well known, and one must turn to manufacturer specifications for the purity and thickness of the beryllium window. Nevertheless, a reasonable estimate of the efficiency can still be obtained, especially if absorption due to the gold and silicon layers is small. This method has been used to compute an efficiency curve for this experiment. Such a curve has also been obtained by Marrus and Schmieder⁴⁴ for a windowless Si(Li) detector in the energy region below 5 keV.

To compute the efficiency curve, we used the following thicknesses: .025 mm $\pm$ 20% Be, dead layer equivalent of .1 micron Si at 6 keV as estimated by the detector manufacturer⁴⁵, and 20 $\mu\text{g}/\text{cm}^2$ Au. Absorption coefficients were taken from the work of Storm and Israel⁴⁰. The results are plotted in Plate XIX along with separate absorption curves for each of the three layers. At energies below 2 keV, the absorption is dominated by the beryllium window, and the correction can be estimated to 20% accuracy as long as the beryllium is pure to better than 99.99%. For krypton L x-rays, the efficiency is estimated to be $49 \pm 10\%$. For x-rays near 3 keV, the absorption is comparable for all three layers. The correction here can be estimated to an accuracy of only 50% because of the uncertainty in the

EXPLANATION OF PLATE XIX

Fractional x-ray transmission through the detector absorbing layers as a function of x-ray energy. Curve 1 is the transmission through 0.1 micron of silicon, curve 2 is that through $20 \mu\text{g}/\text{cm}^2$ of gold, curve 3 is that through .025 mm beryllium, and curve 4 is the total transmission efficiency obtained from the product of curves 1, 2, and 3.

PLATE XIX



thickness of the gold layer. For argon K x-rays, the estimated efficiency is $85 \pm 5\%$.

For the higher energy K x-rays of krypton, the absorption is less than 1%, and the efficiency is taken to be unity. At these energies, the efficiency may be dominated by collection effects within the detector since it was normally operated at a bias voltage of 300 volts. This could be raised to 1000 volts with a slight deterioration of detector resolution but an increase of collection efficiency if higher energy x-rays were to be observed. Because the correction is statistically insignificant for the K x-rays of krypton, this was not done.

DATA ANALYSIS

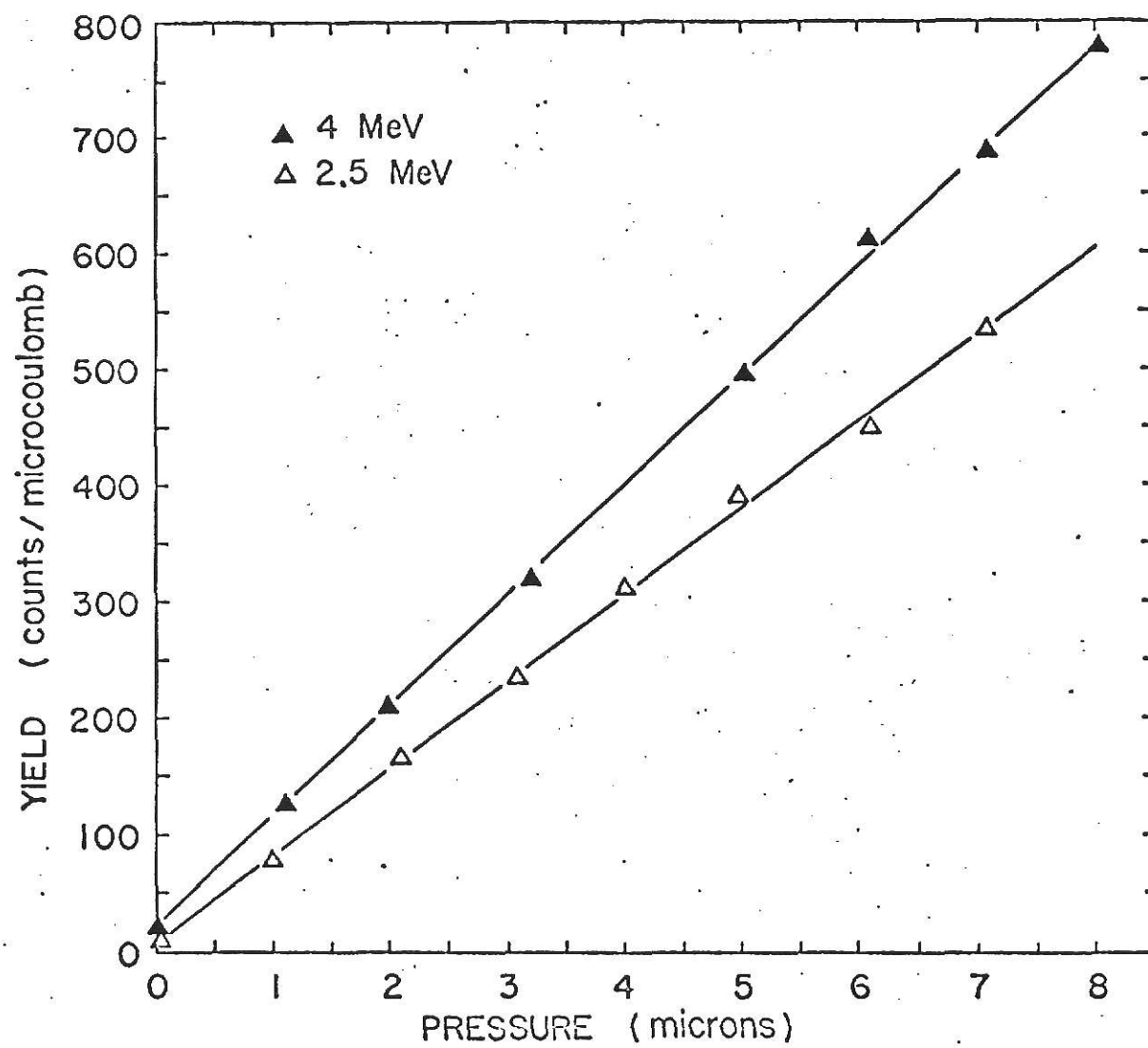
For each target gas at each energy, x-ray yields were obtained for at least five different target thicknesses with manometer pressures from zero to five microns. At each pressure setting, x-ray spectra were obtained for a fixed number of protons monitored by the current integrator during electronic live time of the detection system. Data for these five points were then fitted by a linear least squares fitting routine⁴⁶, and the yield of x-rays was determined per micron of gas pressure on the capacitance manometer. By using this procedure, errors resulting from statistical uncertainty in the x-ray counts and relative uncertainty in the gas pressure were less than 2% for argon K and krypton L x-ray yields and less than 3% for krypton K x-ray yields. Examples of the linear fit for all the lines are shown in Plates XX to XXIII. Separate data points deviate from the fit by less than 2% for krypton L and argon K x-rays and less than 10% for krypton K x-rays. Application of the pressure correction discussed previously to the fitted yields then gave yields normalized to one micron target pressure.

The procedure used to obtain the x-ray yields from the spectra obtained on the multichannel analyzer was to set markers on either side of the x-ray peaks at the points where the peaks merged into the background and then sum the counts channel by channel between the markers. The integration was done either by a PDP-15 computer or from a printout of the data. For argon, the full K x-ray peak was integrated since the $K\beta$ appeared only as a shoulder on the larger $K\alpha$ peak as shown in Plate VIII. For krypton, however, the $K\alpha$ and $K\beta$ peaks were well resolved so that these were integrated separately. The L x-ray peak of krypton showed no signs of discrete lines;

EXPLANATION OF PLATE XX

Yield of argon K x-rays as a function of manometer pressure for incident proton energies of 2.5 (open triangles) and 4.0 (closed triangles) MeV. The yields are normalized to one microcoulomb collected charge and are corrected for electronic dead time.

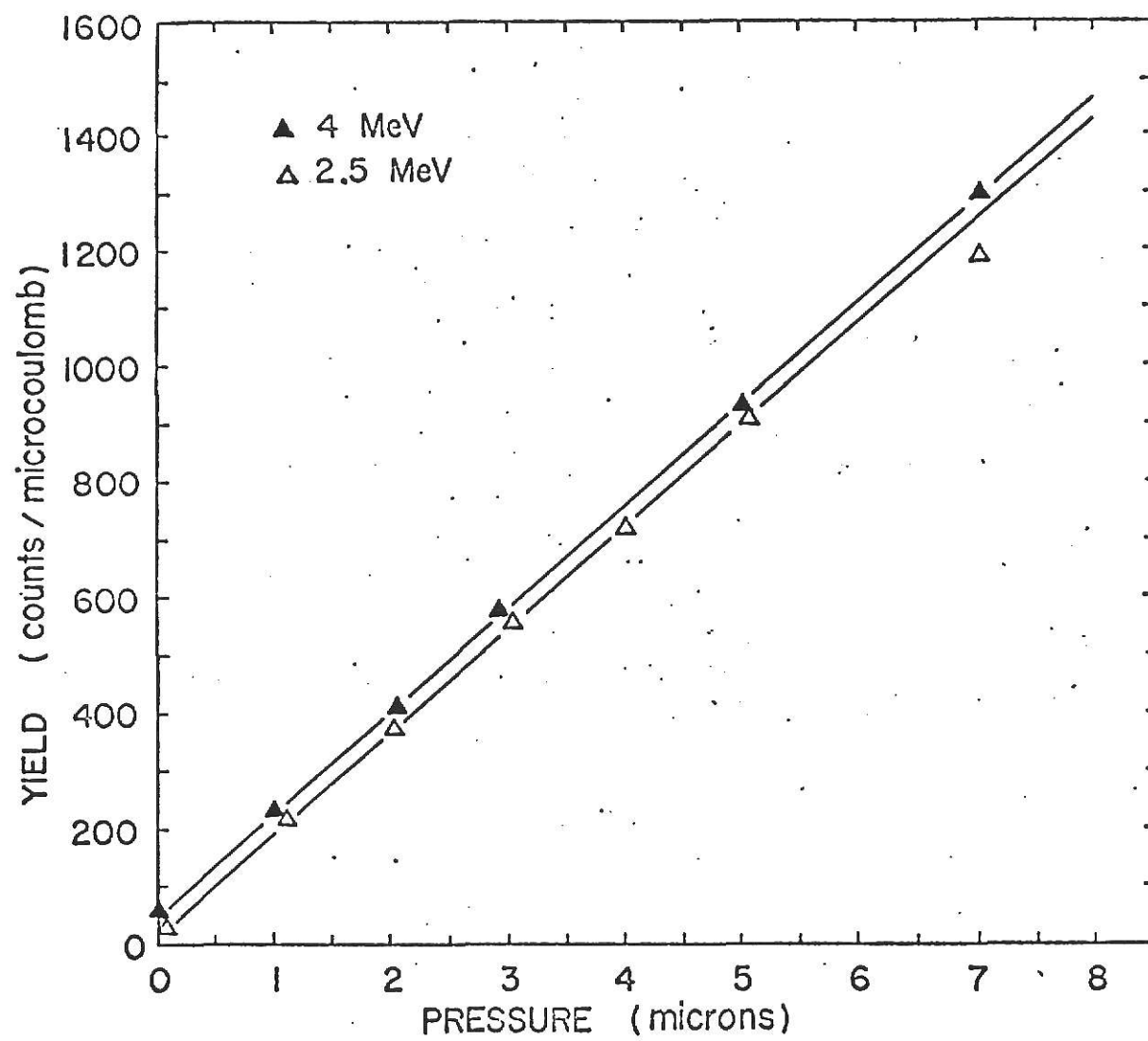
PLATE XX



EXPLANATION OF PLATE XXI

Yield of krypton L x-rays as a function of manometer pressure for incident proton energies of 2.5 (open triangles) and 4.0(closed triangles) MeV. The yields are normalized to one microcoulomb collected charge and are corrected for electronic dead time.

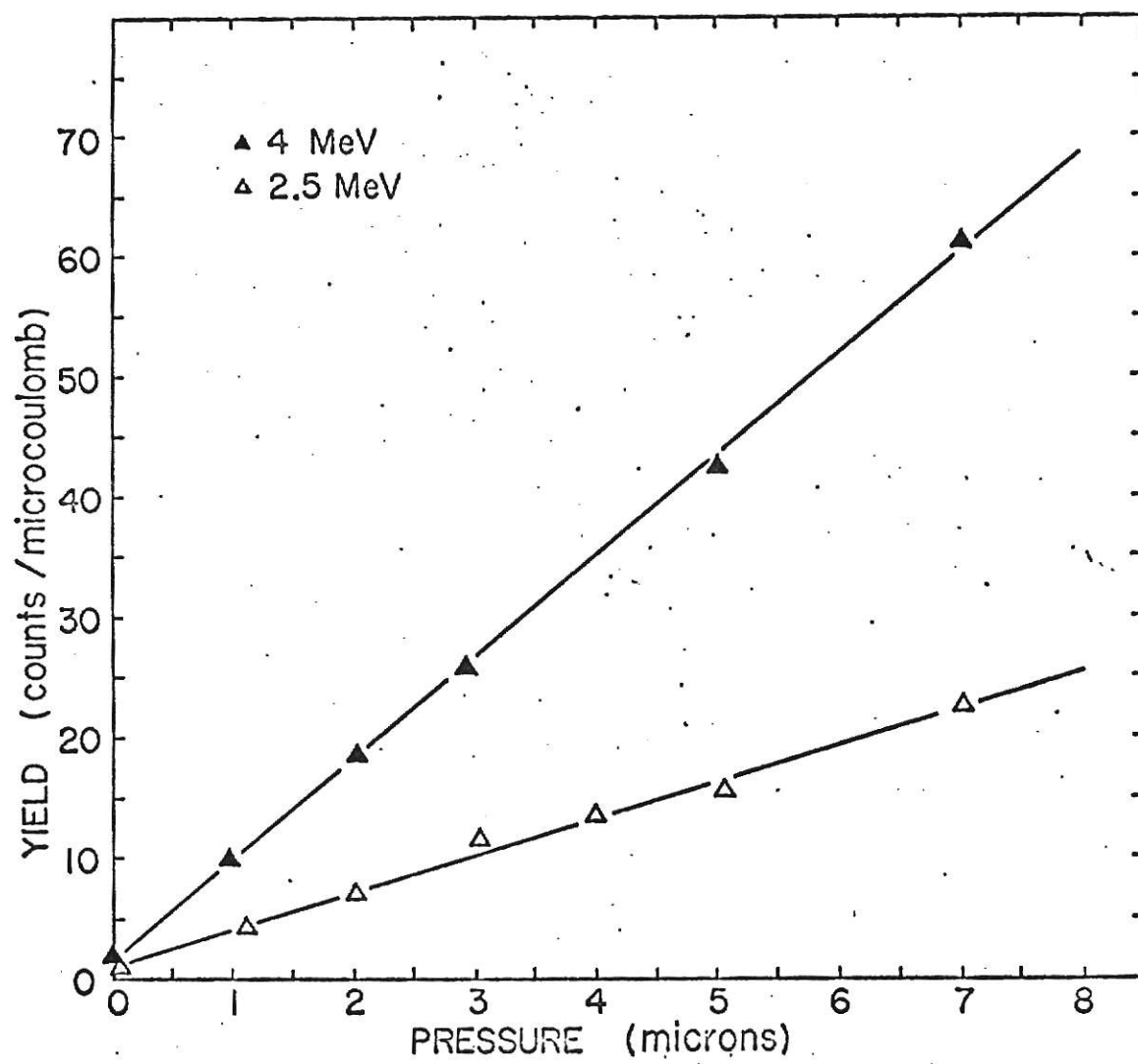
PLATE XXI



EXPLANATION OF PLATE XXII

Yield of krypton $K\alpha$ x-rays as a function of manometer pressure for incident proton energies of 2.5 (open triangles) and 4.0 (closed triangles) MeV. The yields are normalized to one microcoulomb collected charge and are corrected for electronic dead time.

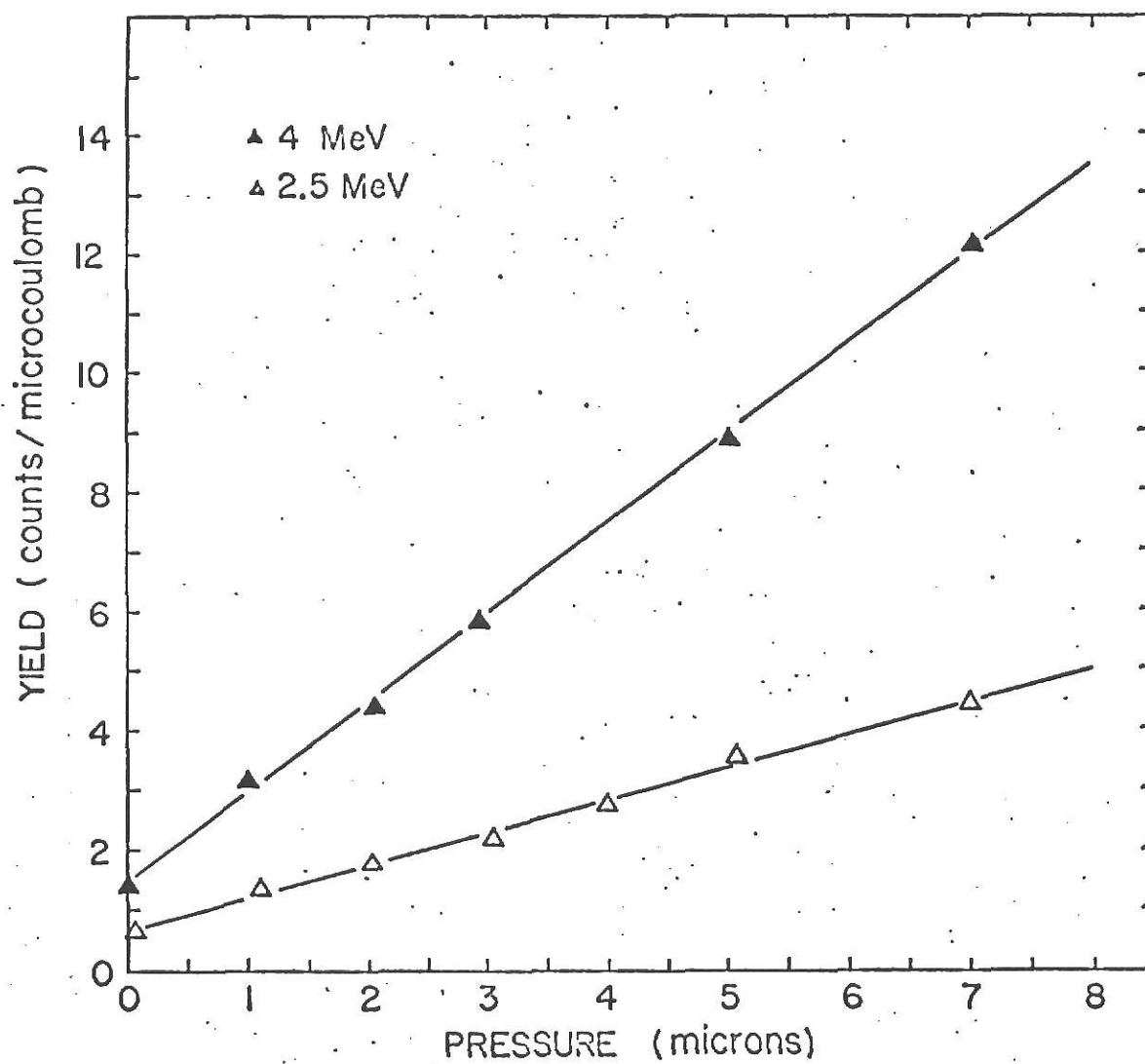
PLATE XXII



EXPLANATION OF PLATE XXIII

Yield of krypton $K\beta$ x-rays as a function of manometer pressure for incident proton energies of 2.5 (open triangles) and 4.0 (closed triangles) MeV. The yields are normalized to one microcoulomb collected charge and are corrected for electronic dead time.

PLATE XXIII



so it, like the argon K, was integrated as a single peak.

In the analysis, no correction was made for a pressure-independent component of background since such a correction was an automatic consequence of the fitting routine discussed above. There was also a pressure-dependent component of background for which no correction was made because its origin was not understood. The contribution of this background to the pressure-normalized x-ray yields is estimated to be less than 5% except at the highest proton energies for which it may be slightly greater. Typical peak-to-background ratios for the three x-ray lines were 80:1 at low proton energy and 40:1 at high proton energy.

An ever-present feature of the argon K and krypton L x-ray lines was the existence of a tail which extended from the low-energy side of the peak to the electronic cut-off. A portion of this feature can be seen in Plate VI for the argon K line and in Plate XIII for the krypton L line. The tail may originate from events related to Compton scattering within the gas cell as well as surface effects on the detector and inefficient charge collection within the detector. To what extent this tail may contain events removed from the full-energy peak has not been determined; however, the total number of counts in the tail amounts to 3% to 10% of the counts in the peak, this percentage being dependent on the proximity of the peak to the electronic cut-off. An experimental efficiency calibration and a study of the detector line shape may help to resolve this problem. No correction was made for it in the present experiment.

Once x-ray yields were obtained, x-ray production cross sections were extracted using the relation

$$\sigma_e = I' / n_1 n \epsilon \Omega_t \quad (3.1)$$

where I' is the x-ray yield corrected for electronic dead time and pressure effects, n is the number of target atoms per unit volume, n_1 is the number of incident protons, $\mathcal{E}$ is the detector efficiency for the x-ray being analyzed, and Ω_t is the total solid angle correction of the detector-gas cell combination. By normalizing I' to one micron target pressure and to one microcoulomb collected charge, equation (3.1) was simplified with $n_1 = 6.25 \times 10^{12}$ protons per μCoul , $n = 3.30 \times 10^{13}$ atoms per cm^3 per micron at 20°C , and $\Omega_t = 1.75 \times 10^{-3} \text{ cm}$ to give

$$\sigma_e = (2.78 \times 10^{-24}) Y_{pq} / \mathcal{E} \quad \text{cm}^2/\text{atom/proton} \quad (3.2)$$

where Y_{pq} is the yield of x-rays per micron of target pressure per microcoulomb of collected charge. Having determined σ_e , ionization cross sections were obtained by dividing σ_e by the fluorescence yield for the particular x-ray as discussed in the introduction.

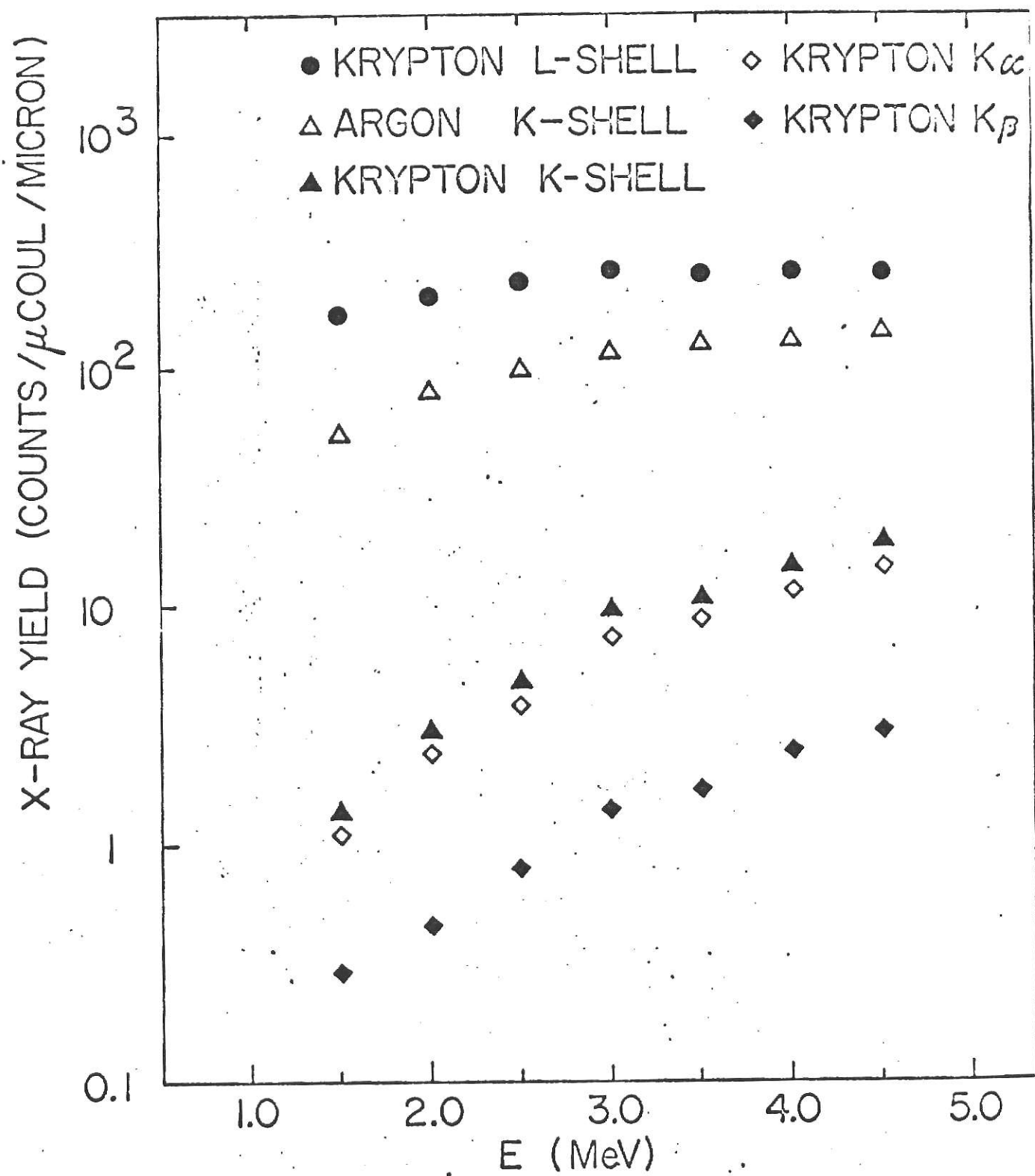
Accumulation of error in quadrature from the data analysis and the various corrections resulted in a total uncertainty of about 20% in absolute cross sections. The relative uncertainty is much better than this since errors due to solid angle, pressure effects, and manometer calibration divide out. For a given x-ray line, the relative uncertainty is about 5%, and for different x-ray lines it is about 10%, the increase being due to the uncertainty in detector efficiency.

The relative uncertainty of 5% for the same x-ray line is borne out by the data. The yield of x-rays as a function of incident proton energy for all the x-ray lines studied is presented in Plate XXIV. These yields are normalized to one micron manometer pressure and one microcoulomb collected charge and are corrected for electronic dead time. For a given x-ray line, the scatter in data points from a smooth curve drawn through the data is

EXPLANATION OF PLATE XXIV

Yield of x-rays as a function of incident proton energy for argon K (open triangles), krypton L (closed circles), krypton K (closed triangles), krypton $K\alpha$ (open diamonds), and krypton $K\beta$ (closed diamonds) x-rays. The yields are normalized to one micron manometer pressure and one microcoulomb collected charge and are corrected for electronic dead time.

PLATE XXIV



typically less than 5%. An exception to this is the krypton data at 3.0 MeV for both the L x-rays and the K x-rays. The deviation for this data is as much as 10% but may be interpreted in terms of the efficiency of beam collection. It is possible that improper beam focussing conditions through the gas cell could result in a significant reduction of incident beam intensity. Thus, for a given amount of collected beam, more x-ray counts would be registered than in the case of total beam transmission. This effect points out the need for taking great care in focussing the beam.

In addition to previously discussed uncertainties, there are a number of possible systematic errors which were not accounted for in the error analysis. Such errors may result from improper gas cell alignment, impurities in or on the beryllium window of the detector, surface effects on the detector, subtraction of background from x-ray peaks, and setting markers for peak integration. The first of these can be kept at a minimum by taking care in the alignment process; however, the others are much more difficult to account for and have not been analyzed in this experiment.

RESULTS AND DISCUSSION

The experimental results are summarized in Tables III and IV. In Table III are listed yields (Y_{pq}), x-ray production cross sections (σ_e), and ionization cross sections (σ_i) for the argon and krypton K-shells and the krypton L-shell. The krypton K-shell results are broken down into separate $K\alpha$ and $K\beta$ yields and x-ray production cross sections in Table IV. Also given are the ratios of $K\alpha$ to $K\beta$ yields. These relative intensities were determined simply to provide a check on the relative accuracy of the data and are not considered to be best values due to uncertainties in detector efficiency and line shape..

For a discussion of the results, see section III of Appendix I.

TABLE III

Experimental X-ray Yields, X-ray Production Cross Sections,
and Ionization Cross Sections

X-ray Line	E (MeV)	Y_{pq} (cts/ μ C/ μ)	σ_e (10^{-24} cm ²)	σ_i (10^{-24} cm ²)
Ar K	1.5	79.3	259.	2180.
	2.0	116.	380.	3190.
	2.5	147.	478.	4020.
	3.0	173.	566.	4750.
	3.5	192.	625.	5250.
	4.0	195.	635.	5340.
	4.5	214.	700.	5880.
Kr K	1.5	2.08	5.78	8.56
	2.0	4.48	12.4	18.4
	2.5	7.44	20.6	30.5
	3.0	13.0	36.1	53.5
	3.5	16.2	45.0	66.7
	4.0	21.8	60.5	89.6
	4.5	27.5	76.4	113.
Kr L	1.5	254.	1440.	60400.
	2.0	309.	1750.	73400.
	2.5	342.	1940.	81400.
	3.0	383.	2170.	91100.
	3.5	368.	2080.	87500.
	4.0	382.	2160.	90800.
	4.5	377.	2135.	89700.

TABLE IV

Experimental X-ray Yields, X-ray Production Cross Sections,
and Relative Intensities for Krypton $K\alpha$ and $K\beta$ X-rays

E (MeV)	Krypton $K\alpha$		Krypton $K\beta$		Rel. In. ($K\alpha/K\beta$)
	Y_{pq} (cts/ $\mu\text{C}/\mu$)	σ_e (10^{-24} cm ²)	Y_{pq} (cts/ $\mu\text{C}/\mu$)	σ_e (10^{-24} cm ²)	
1.5	1.64	4.56	.441	1.22	3.72
2.0	3.80	10.5	.684	1.90	5.56
2.5	6.11	17.0	1.19	3.29	5.13
3.0	10.9	30.3	2.10	5.82	5.19
3.5	13.7	37.9	2.55	7.09	5.37
4.0	18.1	50.2	3.71	10.3	4.88
4.5	23.1	64.0	4.48	12.4	5.16

APPENDIX I

"Experimental Determination of Cross Sections for the
Production of X-rays of Argon and Krypton by 1.5 to 4.0 MeV Protons"

Experimental Determination of Cross Sections for the
Production of X-rays of Argon and Krypton by 1.5 to 4.0 MeV Protons.*

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I. Introduction

The process of inner shell ionization in proton-atom collisions has been described by Merzbacher and Lewis¹ in terms of a plane wave Born approximation (PWBA). More recently, Garcia² has proposed a classical binary-encounter model (BEM), which he shows to be equivalent to an impulse approximation. Both the PWBA and BEM treatments predict an excitation function which rises steeply from the origin to a broad maximum and then falls off gradually. However, for incident proton energies in the vicinity of this maximum, the BEM predicts larger cross sections than the PWBA and is in slightly better agreement with the experimental data.³ In these experiments, the ionization phenomena are studied indirectly by observing characteristic X-rays emitted in the de-excitation process. Most of the work has been done using solid targets, although Kessel, *et. al.*⁴ have observed K X-rays of argon in collisions with 3 MeV protons while Saris and Onderdelinden⁵ have obtained argon L X-ray yields for proton energies of 70 to 130 keV. By using gaseous rather than solid targets, one may achieve negligible self-absorption of X-rays in the target as well as single collision conditions, thus making the gas target a potentially more powerful tool for the study of the collisional mechanisms. In the present experiment, thin targets of argon and krypton were bombarded by 1.5 to 4.5 MeV protons, and K and L X-rays were observed. Absolute X-ray production cross sections as well as ionization cross sections were determined and, in the case of K-shell ionization, compared with the PWBA and BEM treatments. For L-shell ionization of krypton, comparison was made with the PWBA description only. In this case, experimental ionization cross sections are quite

sensitive to the choice of fluorescence yield.

II. Experimental Methods

A schematic diagram of the experimental apparatus used to observe X-ray production in gases is shown in Fig. 1. A momentum-analyzed proton beam from the Tandem Van de Graaff accelerator passes through the differentially pumped gas cell and is collected by a suppressed Faraday cup and integrated in a Brookhaven current integrator. Beam currents ranged from 500 na to 1 ua.

X-rays resulting from collisions in the gas cell are detected by a Si(Li) detector with an active area of 80 mm^2 mounted inside the gas cell at right angles to the beam direction and at a distance of 7.66 cm from the beam. The interaction region of the gas cell viewed by the detector is 1.70 cm in length and the solid angle used to calculate total X-ray yields is .0136 steradian. To reduce background radiation, the gas cell is lined with graphite inserts; in addition, the beam apertures of the cell are fabricated from graphite. The detector is energy calibrated by means of a removable iron-55 source mounted within the gas cell. The energy resolution on the 5.9 keV MnK α line is typically about 200 eV FWHM. A pulsed optical feedback preamplifier is used with the detector and, as a result, a significant dead time correction to X-ray yields must be made. This is done electronically by using the amplifier gating pulse to count integrated beam current for detector live time only. Tests performed with an X-ray source and two amplification systems give an accuracy of 2% to this correction technique even under high counting rate conditions for which the dead time is unusually large. Under beam conditions of this experiment, the correction was less than 40% at the highest counting rates.

A major factor in determining absolute yields of low energy X-rays is to account correctly for absorption between source and detector. One distinct advantage in a gas target experiment is that self-absorption is extremely small; for example, it amounts to less than 0.1% for krypton L X-rays in our geometry at a pressure of 10 microns. To account for absorption in the .0025 cm beryllium window and the gold and silicon dead layer of the detector, an efficiency curve as a function of X-ray energy was estimated from absorption coefficients of Storm and Israel.⁶ At 6 KeV, the detector manufacturer⁷ estimates a dead layer equivalent to 0.1 micron of silicon for our detector. We have used this amount plus $40 \mu\text{g}/\text{cm}^2$ of gold to obtain the efficiency curve. In the region below 2 keV, the total absorption is dominated by the effect of the beryllium window, and the correction can be estimated to 20% accuracy. For krypton L X-rays (1.63 keV), the estimated efficiency is $51 \pm 10\%$. For X-rays near 3 keV, the absorption is comparable for the beryllium, gold, and silicon layers, and the correction can be estimated to no better than 50% because of the uncertainty in the thickness of the gold layer. For argon K X-rays (2.97 keV), the estimated efficiency is $89 \pm 5\%$. For the higher energy K X-rays of krypton ($K\alpha$ at 12.63 keV and $K\beta$ at 14.13 keV), the absorption correction is very small and is probably dominated in determining the efficiency by the onset of collection effects within the detector.

The pressure in the gas cell was determined by means of an MKS Baratron capacitance manometer coupled to a Granville-Phillips leak valve for controlling the target thickness. However, the pressure was not regulated while X-ray data was accumulating since vibrations produced by the pressure

controller considerably degraded the energy resolution of the highly microphonic detector.

The absolute accuracy of the manometer calibration is less than 10% and the precision of the pressure setting was considerably better than 5%. In order to obtain complete beam transmission through the gas targets, exit apertures with diameters of 2 mm and 3 mm were used on the cell. The flow of gas through these large apertures required a significant correction to the target thickness because of the pressure gradient between the manometer and the cell. It can be shown that the target pressure P_T is related to the manometer pressure P_M by the relation

$$P_T = P_M (1 + bd^2)^{-1}$$

where d is the diameter of the apertures and b is independent of the target gas. Using five different sets of apertures the magnitude of the correction was obtained to an accuracy of 15%. For the data obtained in this experiment, corrections of up to $50 \pm 7\%$ have been applied.

Although the gas cell was normally operated at pressures below five microns, thin target conditions were found to prevail up to manometer pressures of at least 30 microns corresponding to a target pressure of 23 microns. This is illustrated in Fig. 2 by the linearity of the yield of krypton L X-rays as a function of manometer pressure. Absolute X-ray production cross sections were extracted from the data by first determining the slopes of the yield versus pressure curves with a linear least squares fitting routine. The slopes were then corrected for target thickness, detector efficiency, target geometry, beam collection, and dead time of the electronic equipment. This procedure resulted in negligible statistical

errors. In addition, it eliminated the need to correct for the pressure independent background. No correction was made for the pressure dependent background, but this is estimated to result in an uncertainty of less than 3% in the slopes. The accumulation of error in quadrature from the experimental uncertainties and the data analysis provides an accuracy of about 20% in absolute X-ray yields and considerably better than that relatively. This figure includes, in addition to previously discussed uncertainties, an 8% uncertainty in the target geometry.

III. Results and Discussion

Analysis of the yield of X-rays as a function of gas target thickness and beam intensity has been used to extract absolute cross sections for the production of X-rays per target atom. Ionization cross sections have been obtained by dividing the cross section for X-ray production by the fluorescence yield. K-shell fluorescence yields used were .119 for argon from the experimental work of Bailey and Swedlund⁸ and .675 for krypton from theoretical calculations of Walters and Bhalla.⁹ Uncertainties introduced by the use of K-shell fluorescence yields are usually small with differences between experimental and theoretical values being less than 5% for the gases considered here. For L-shell ionization of krypton, however, the situation is quite different. The authors are aware of only two experimental measurements of the krypton L-shell fluorescence yield.¹⁰ Recent theoretical calculations of McGuire¹¹ differ from the experimental values by factors of 3 and 6. After including the contribution to the fluorescence yield for Coster-Kronig transitions that also have been calculated by McGuire, ionization cross sections are obtained which are larger than those obtained using the older experimental values, but are in considerably better agreement with experimental ionization measurements. The mean krypton L-shell fluorescence yield of .0238 derived from McGuire's work was chosen for the data in this experiment. No estimate of error is given, although it may be considerable.

The experimental K-shell ionization cross sections of krypton and argon that have been obtained for protons in the energy region from 1.5 to 4.5 MeV are shown in Fig. 3. Also included in the figure are theoretical

calculations of these cross sections in the PWBA and BEM approximations.^{12,3} As suggested by Garcia,² the incident energy, E , and the cross section, σ_k , have been scaled with the K-shell binding energy, I_k , to provide a universal theoretical curve for comparison with experiment.

For krypton, the experimental points are consistently higher than the theoretical curves, with absolute agreement within 25% for the PWBA and a factor of two for the BEM. The energy dependence of the experimental data compares favorably with both approximations in the lower energy range, but at the upper end of the interval the data may slightly favor the BEM over the PWBA which rises less steeply in the region below the peak.

The energy region of the experiment for argon K X-rays is considerably closer to the peak of the theoretical ionization cross section than for K X-rays of krypton. In this region, the results of the two approximations cross over while the energy dependence of the experimental data is intermediate, rising more steeply than the PWBA and less so than the BEM. In absolute value, the argon K-shell ionization cross sections are lower than the theoretical results by amounts ranging from about 5% to over 30%.

Although the absolute experimental uncertainty of the ionization cross sections is of the same order as the discrepancy with the theoretical models, the relative uncertainty is considerably less than this between the two target gases for which the solid angle, pressure corrections, and electronic dead time effects are the same. It is not expected that detector efficiency and fluorescence yields can account fully for the discrepancy with theoretical models in opposite directions for the argon and krypton data.

L-shell ionization cross sections of krypton by protons from 1.5 to 4.5 MeV are presented in Fig. 4 and compared to the results of a calculation in the Born approximation. The scaling adopted in presenting the data is that of Merzbacher and Lewis.¹ The dimensionless parameter used for the energy dependence is $\eta_L = \frac{mE}{MZ_L^2 (\text{Rydberg})}$ where m/M is the ratio of electron to proton mass and Z_L is the screened nuclear charge seen by an L-shell electron. Agreement between theory and experiment is quite favorable with both exhibiting approach to a broad maximum. The absolute agreement is within 10% but must be considered somewhat fortuitous because of the magnitude of the experimental uncertainty and the fluorescence yield.

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Figure Captions

- Figure 1 Experimental apparatus for the determination of X-ray yields from gases.
- Figure 2 Yield of krypton L X-rays as a function of manometer pressure.
- Figure 3 K-shell ionization cross sections as a function of incident proton energy. The open and solid triangles are the experimental results for argon and krypton respectively. The solid curve is the Born approximation result¹² and the dashed curve is the impulse approximation result of Garcia's binary encounter model.³
- Figure 4 L-shell ionization cross sections for krypton as a function of the dimensionless energy parameter η_L . The closed circles are the experimental results, and the solid line is the Born approximation result.¹²

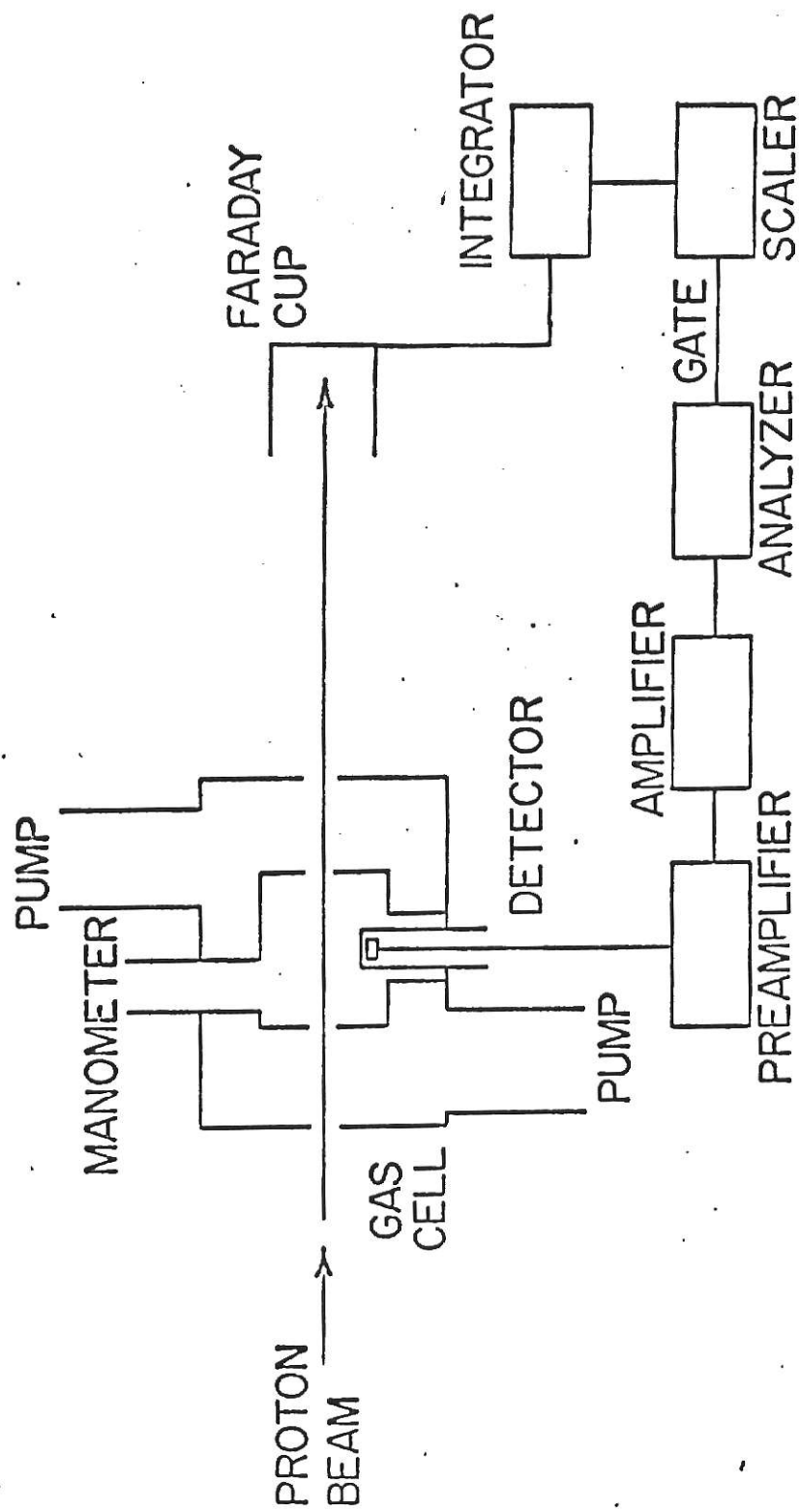


FIGURE 1

FIGURE 2

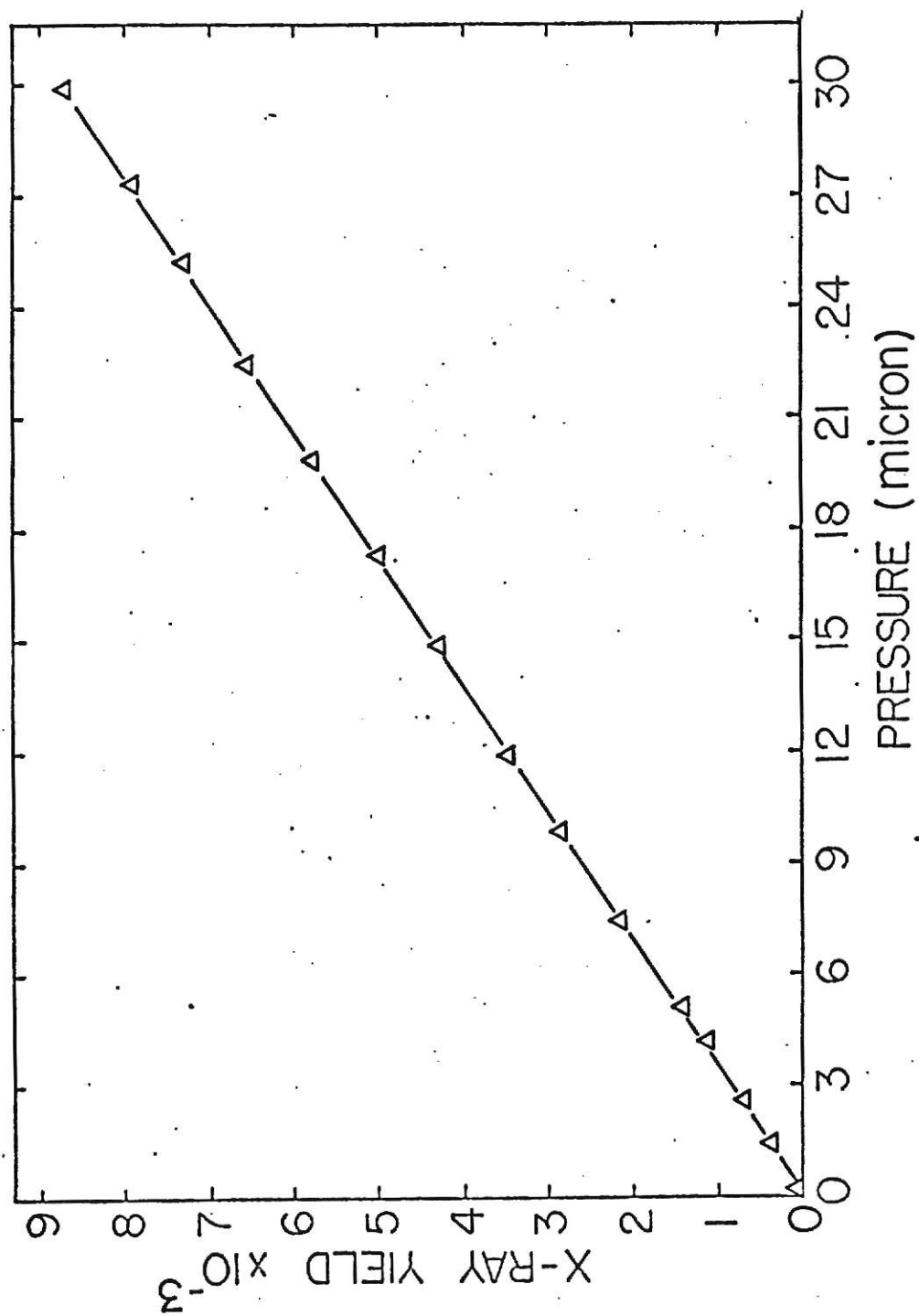


FIGURE 3

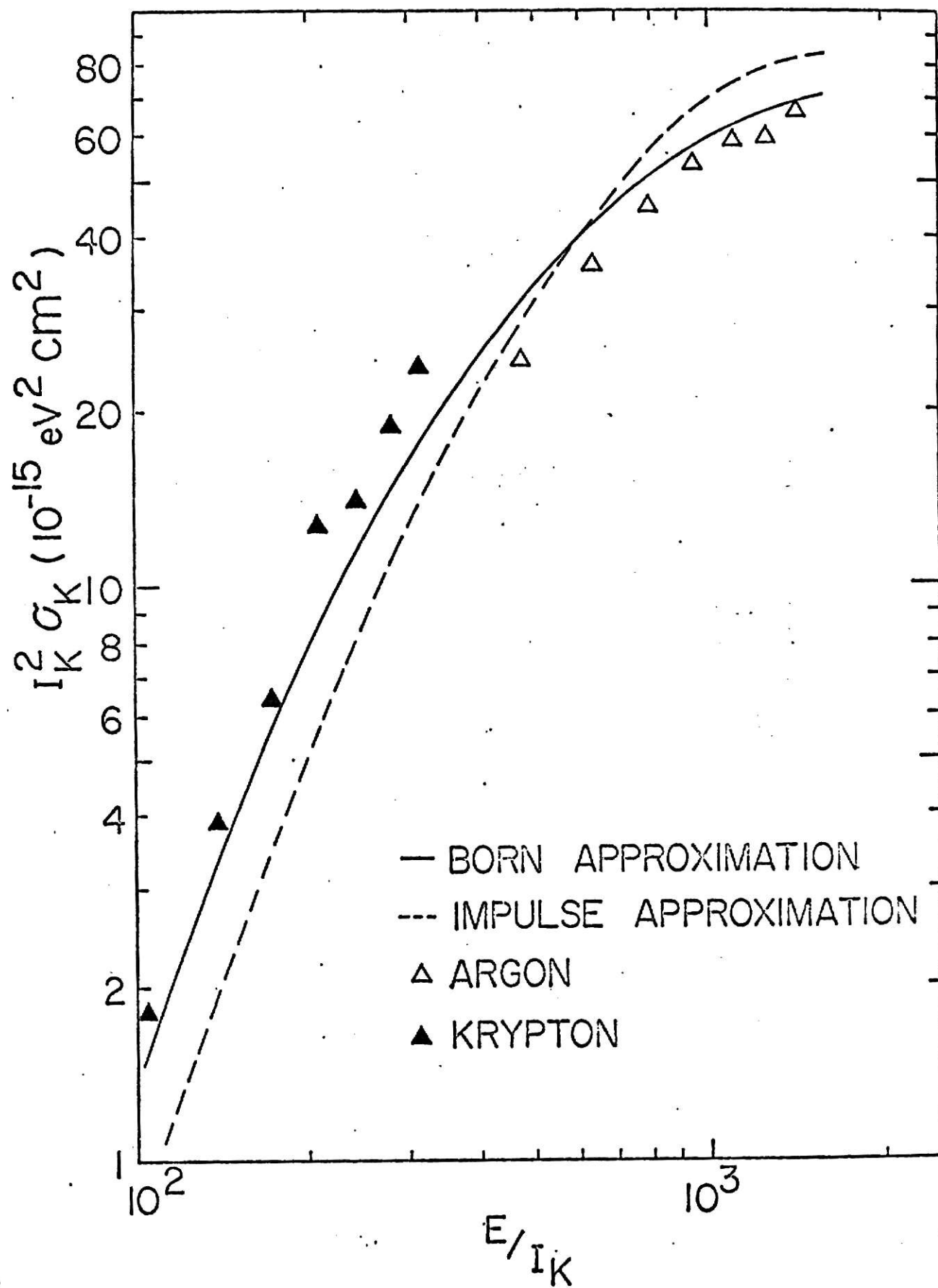
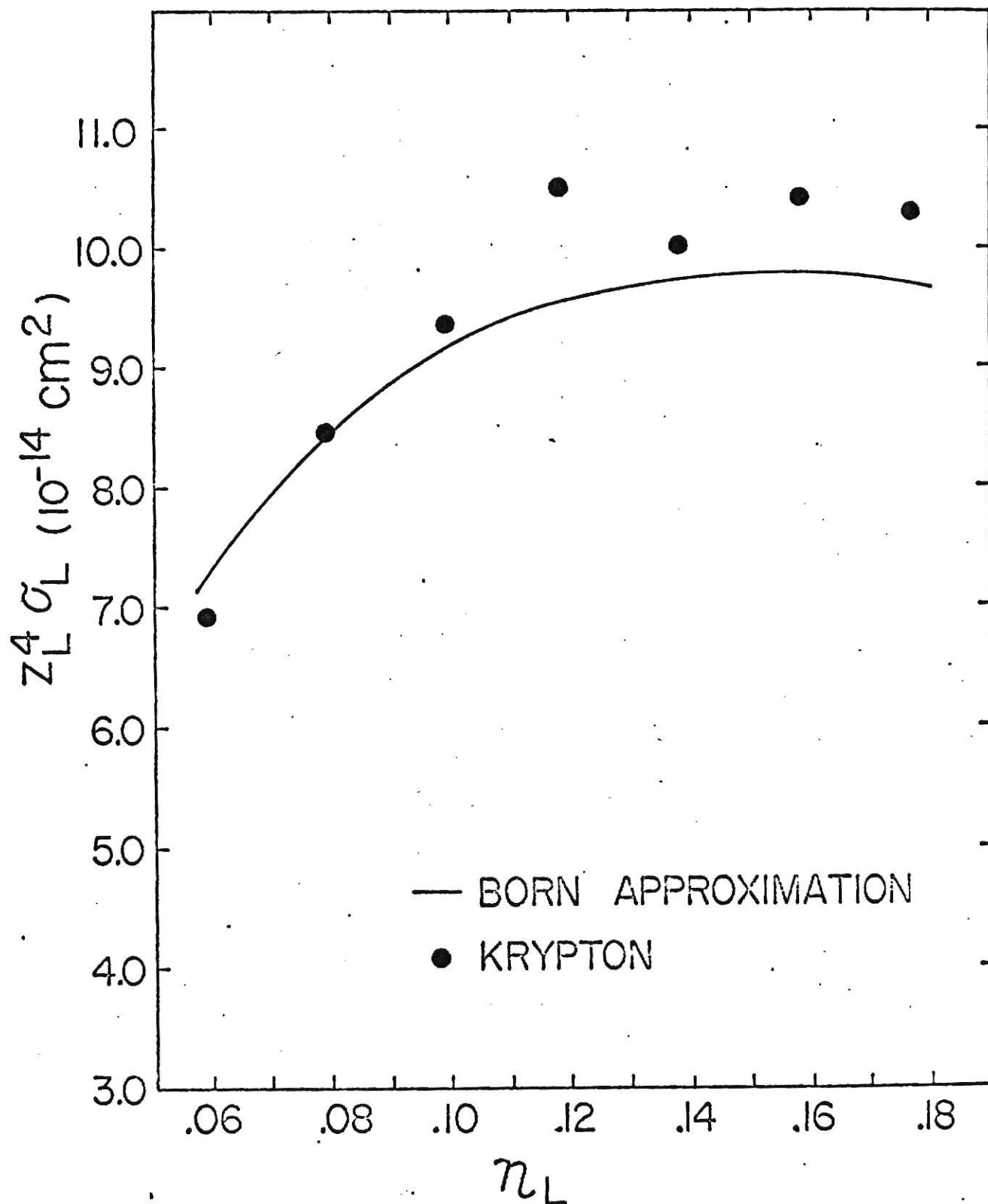


FIGURE 4



APPENDIX II

Inner Shell Ionization of Xenon by Proton Bombardment

(Preliminary Results)

Herein is reported the initial step in an extension of the present experiment to proton-induced ionization of xenon. The experimental setup for the xenon was exactly the same as that reported previously with the exception that the inner gas-cell apertures each had a diameter of 1.5 mm. Xenon L x-ray yields were obtained for manometer pressures of zero to fifteen microns at a single proton energy--3.0 MeV. These yields have not yet been converted to cross sections since a method of data analysis for the complicated spectra has so far not been developed by the author.

The L x-ray spectrum is shown in Plate XXV. By using an experimental x-ray energy calibration and x-ray energies of Storm and Israel⁴⁰, the major components of each of the six x-ray peaks were determined. These components are given in Table V with the energy of the midpoint of each peak. In addition, the particular transitions associated with the various lines are listed in Table VI.

No K x-rays were observed, this probably being the result of the very small K-shell ionization cross section of xenon. Using higher energy proton beams may reveal the presence of these x-rays.

This is the extent of the present work with xenon. The next step will be to obtain an L-shell excitation function for proton energies between one and five MeV.

EXPLANATION OF PLATE XXV

Pulse height spectrum of the xenon L x-rays. The incident proton energy is 3.0 MeV.

PLATE XXV

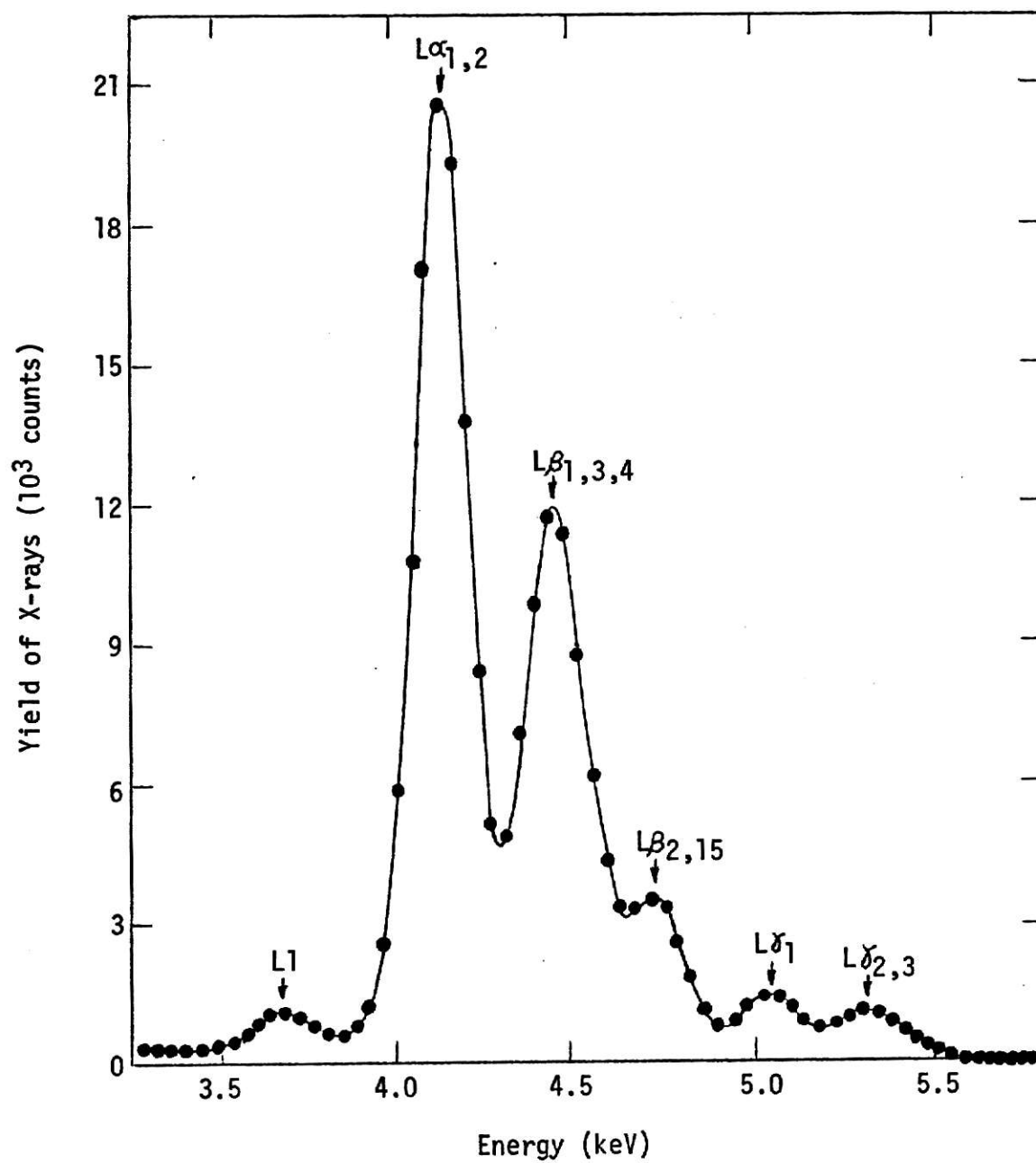


TABLE V

Calibration of the Xenon L X-ray Spectrum

X-ray Lines	Experimental X-ray Energy (keV)
L _I	3.705
L $\alpha_{1,2}$	4.132
L $\beta_{1,3,4}$	4.447
L $\beta_{2,15}$	4.698
L γ_1	5.027
L $\gamma_{2,3}$	5.279

TABLE VI

X-ray Notation

X-ray Notation	Transition
K α	K-L
K β	K-M,N
L α_1	L _{III} -M _V
L α_2	L _{III} -M _{IV}
L β_1	L _{II} -M _{IV}
L β_2	L _{III} -N _V
L β_3	L _I -M _{III}
L β_4	L _I -M _{II}
L β_{15}	L _{III} -N _{IV}
L γ_1	L _{II} -N _{IV}
L γ_2	L _I -N _{II}
L γ_3	L _I -N _{III}
L _I	L _{III} -M _I
L γ	L _{II} -M _I

LIST OF SYMBOLS

<u>Symbol</u>	<u>Description</u>
v	Velocity of the incident proton
u	Effective velocity of an atomic electron in its orbit
m	Mass of the electron
M	Mass of the proton
T_m	Maximum energy which one particle can transfer to another particle in a free collision
s	Principal quantum number as indicated by $s = K (1), L (2), \dots$
I_s	Actual ionization potential of the s-shell
E	Incident proton energy
z	Charge of the projectile
Z	Actual charge of the atomic nucleus
Z_s	Screened nuclear charge as seen by an s-shell electron
η_s	mE/MZ_s^2 (Rydberg), the energy-scaling parameter in the Born approximation
θ_s	$I_s s^2 / Z_s^2$ (Rydberg), the screening parameter in the Born approximation
σ_s	Cross section for ionization of the atomic s-shell in an ion-atom collision
σ_i	Generalized notation for the ionization cross section
σ_e	Cross section for production of characteristic x-rays of the target atom in a proton-atom collision
ω_s	Fluorescence yield of the s-shell
ω_i	Fluorescence yield of the i subshell
a_i	Auger yield of the i subshell
f_{ij}	Coster-Kronig yield corresponding to the transition L_i to L_j
γ_i	Generalized fluorescence yield for the i subshell (includes the effect of Coster-Kronig transitions)

$\bar{Y}_L$	Generalized mean L-shell fluorescence yield
n	Number density of target atoms
n_i	Number of protons incident on the target
$\mathcal{E}$	Efficiency of the x-ray detector for a given x-ray energy
P_M	Pressure indicated by the capacitance manometer
P_T	Pressure of the target gas
I	Observed yield of x-rays normalized to one microcoulomb collected charge and corrected for electronic dead time
Y	Slope of the plot of I versus P_M
Y_{pq}	Slope of the plot of I versus P_T
d	Diameter of an inner gas cell aperture
x	Distance from the proton beam to the detector
A	Active area of the detector
L	Interaction length seen by all points of the detector
l	Interaction length seen by fractional portions of the detector
L'	Total interaction length viewed by the detector ($L + 2l$)

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INNER SHELL IONIZATION OF NOBLE
GAS ATOMS BY PROTON BOMBARDMENT

by

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ABSTRACT

Thin gas targets of argon and krypton were bombarded by protons of 1.5 to 4.5 MeV energy, and the resulting x-radiation was observed with a Si(Li) detector. K-shell x-rays of argon and krypton and L-shell x-rays of krypton were identified, and the total yields of x-rays as a function of gas target thickness in the region of zero to five microns were determined for each of the three cases. For both gases, the $K\alpha$ and $K\beta$ components were observed; however, separate x-ray yields for these components were determined only for the case of krypton. Absolute cross sections for the production of K- and L-shell x-rays were obtained and converted to ionization cross sections by using recent theoretical and experimental values of the fluorescence yields. The K-shell ionization cross sections were compared to the theoretical predictions of the Born approximation as well as to the binary-encounter model of Garcia. For krypton, the experimental results are consistently higher than both theoretical results while the reverse is true for argon. However, the energy dependence of the argon cross sections, which fall near the peak of the excitation function, is intermediate to the two theoretical predictions. For krypton, this energy dependence agrees well with both theories except at the higher collision energies for which the binary-encounter model may be favored slightly. The L-shell ionization cross sections were compared only to the Born approximation, with agreement being favorable. These experimental L-shell cross sections are sensitive to the choice of fluorescence yield for which few experimental values are available, and this results in considerable experimental uncertainty.