SPATIAL DISTRIBUTION OF IONIZATION ENERGY, AND TRACK WIDTH

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B. S., Kansas State University, 1965

A MASTER'S THESIS

submitted in partial fulfillment of the

requirements for the degree

MASTER OF SCIENCE

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1967

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INTRODUCTION

Due to the availability of accelerators, such as those at Yale and Berkeley, capable of accelerating ions of charge up to 18e with energies up to 10 MeV per nucleon, and the ability of balloons and rockets to reach the outer limits of the earth's atmosphere where heavy ions with energies up to 10^{10} eV are found in the primary cosmic radiation, scientists have been interested in obtaining knowledge of the interactions of heavy ions with matter and of the practical application of these interactions.

In the process of interacting with matter, a heavy ion transfers its energy to the surrounding medium. Therefore, it is reasonable to assume that several phenomena, such as the width of ion and monopole tracks in emulsion, radiation damage to biological materials and to solids, and related processes, can be explained by means of an accurate description of the way in which the ion's energy is transferred to the surrounding medium.

It was the purpose of the research that is reported in this thesis to calculate the spatial distribution of ionization energy associated with a heavy ion and to use the results as a means of describing the width of ion tracks in emulsion.

SPATIAL DISTRIBUTION OF IONIZATION ENERGY

As a heavy ion passes through matter, it loses energy by ionization, that is, the ion ejects electrons, commonly called delta rays, out of the atoms of the medium. As these primary delta rays scatter out from the ion's path, they in turn lose their energy through interactions with other electrons of the medium.

By the spatial distribution of ionization energy, we mean the energy density, E^+, given as a function of radial distance, x, from the ion's path, which is deposited by delta rays within a cylindrical shell of unit length and mean radius x centered on the ion's path. We can write an expression for the density of energy deposited as

$$E^+ = -1/(2\pi x)(\partial E/\partial x), \tag{1}$$

where E is the total energy carried through a cylindrical surface of radius x by delta rays. To find the energy E, we integrate the product of W, the energy carried outside the cylinder by a single delta ray with initial energy w, and σ, the number of delta rays with initial energy w that penetrate the cylinder, over the appropriate energies. Since only a fraction, η, of the delta rays of initial energy w pass through a cylinder of given radius, we write the quantity, σ, as ηdn, where dn is the initial number of delta rays with initial energies between w and w + dw generated within the cylinder by the passing ion and η is the fraction of the initial number of delta rays with initial energy w that penetrate the cylinder of radius x. This symbolic structure is dictated by available data on the penetration of electrons through matter. In these terms our expression for E and E^+ are

$$E = \int_{W_x}^{W_{max}} W\eta(dn/dw)dw, \tag{2}$$

and

$$E^+ = \left[-1/(2\pi x)\right](\partial\ /\partial x)\left(\int_{W_x}^{W_{max}} W\eta(dn/dw)dw\right), \tag{3}$$

where W_{max} is the maximum energy that can be given to an electron by a passing ion and W_x is the maximum initial energy that an electron which just reaches the surface of the cylinder can have.

By use of the chain rule for partial differentiation Eq. (3) becomes

$$E^{+} = \left[-1/(2\pi x)\right](\partial W_x/\partial x)(\partial\ /\partial W_x)\left(\int_{W_x}^{W_{max}} W\eta(dn/dw)dw\right). \tag{4}$$

Certain results from theory and from experiment must be used to evaluate Eq. (4).

The expression for dn/dw is given by the classical delta ray distribution formula

$$dn/dw = (2\pi e^4 NZ'^2)/(mc'^2\beta^2 w^2), \tag{5}$$

where e and m are the charge and mass of an electron, N is the electron density of the medium, c' is the speed of light in vacuum, β is the ratio of the speed of the ion to the speed of light, and Z'e is the effective charge of the ion.

An empirical expression for Z' as given by Barkas[1] is

$$Z'e = Ze\left[1 - \exp(-125\beta/Z^{2/3})\right], \tag{6}$$

where Z is the atomic number of the ion. This expression for Z' takes into account electron capture by the ion as it nears the end of its range, and is based on studies of the range of machine accelerated ions in emulsion.

The expression for W_{max} is determined by kinematics, if the mass of the ion is much greater than the mass of the electron, as

$$W_{max} = 2mc'^2\beta^2/(1 - \beta^2). \tag{7}$$

Studies of the penetration of normally incident electrons through thin films of various materials, most commonly aluminum, were carried out by Kanter and Sternglass[2], for electrons in the energy interval of 0.6 - 10.0 keV, by Cosslett and Thomas[3], for the energy interval of 2.0 - 15.0 keV, and by Katz and Penfold[4], for the energy interval 10.0 keV - 20.0 Mev. The information presented in these papers was combined in a range-energy relaion for electrons suggest by Weber[5],

$$r = Aw[1 - B/(1 + Cw)], \tag{8a}$$

$$w = \{AB - A + rC + [r^2C^2 + 2AC(B + 1)r + (AB - A)^2]^{\frac{1}{2}}\}/(2AC), \tag{8b}$$

where r is the practical range of an electron with initial energy w. The constants A, B, and C were determined by a least squares fit to this information for aluminum, to be $A = 4.468 \times 10^4$ grams$\cdot$(cm$^2\cdot$ergs)$^{-1}$, B = 0.9824, and $C = 1.771 \times 10^6$ ergs^{-1}. With these values for the constants A, B, and C, Eq. (8a) fits the available information to within about 3% over the energy interval of 0.6 keV - 20.0 MeV. When expressed in units of grams/cm^2, the practical range of an electron is independent of the atomic number of the medium. To express the practical range in centimeters, the constant A must be divided by the density of the medium, ρ. The range-energy relation computed from Eq. (8a) is plotted in Plate I.

According to Kanter[6], the fractional number, η, of normally incident electrons of energy w transmitted through a film of thickness r_o may be represented as a function of w/W_{r_o}, where W_{r_o} is the initial energy required for an electron to traverse a distance r_o. The form,

$$\eta = 1 - \exp[G(w/W_{r_o} - 1)], \tag{9}$$

with G = -1.263 fits Kanter's data to within about 3%. We extrapolate the application of Eq. (9) to all energies and thicknesses in the present work.

The information used in obtaining Eqs. (8) and (9) was acquired for electrons normally incident onto plane films. In the application of Eqs.

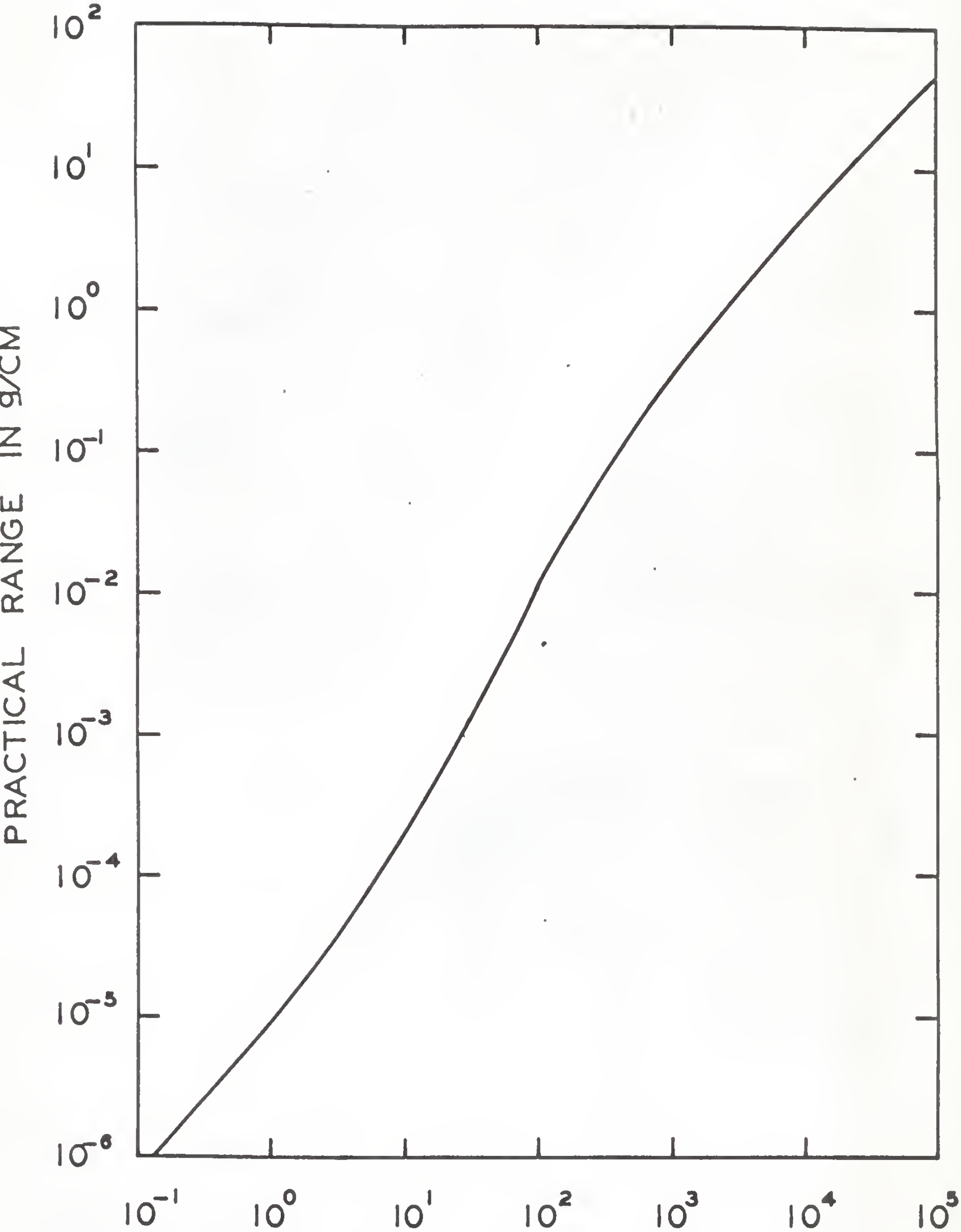
EXPLANATION OF PLATE I

Plot of the Pratical Range

Versus Initial Energy for

Electrons in Aluminum.

PLATE I


PRACTICAL RANGE IN g/CM^2
10^2
10^1
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-1}
10^0
10^1
10^2
10^3
10^4
10^5
INITIAL ENERGY IN KEV

(8) and (9) to the problem of determining the spatial distribution of ionization energy, the difference between electrons normally incident onto plane films and delta rays radially ejected from the axis of a cylinder is ignored. Through the use of Eqs. (8) and (9), problems of delta ray scattering and secondary delta ray production are automatically accomodated.

To the lowest order of approximation, we may take electrons to move in straight paths. If the range of an electron of initial energy w is given by r, then the residual energy, W, of an electron which has gone a distance r_o is given by

$$W(r) = w(r - r_o). \tag{10}$$

To be able to get W and W_x as functions of the radial distance, x, from the ion's path, the relation between x and the linear distance traversed, r, must be found.

If the electrons of the medium can be considered free, the angle of ejection, θ, of a delta ray with respect to the ion's path is uniquely determined by its initial energy. The angle, θ, as determined by kinematics is

$$\theta = \cos^{-1}[w/(2mc'^2\beta^2 + w\beta^2)]^{\frac{1}{2}}. \tag{11}$$

Thus the relation between x and r_o is

$$x = r_o \sin\theta, \tag{12}$$

where r_o is the range of a delta ray that ejected at an angle θ will just travel to the cylinder's surface.

The electrons of the medium are not free; therefore, the angular distribution of delta rays is not uniquely known. Because the electrons are bound to the atoms of the medium and because of delta ray scattering, it is assumed that the delta rays are totally diffuse; that is, they follow a $\cos\theta$ distribution. On the average, the relation between x and r_o now becomes

$$x_{ave.} = 0.5r_o, \tag{13}$$

where the factor of 0.5 is the average of $\cos\theta$ over a hemishpere.

In the subsequent discussion, we will drop the subscript ave.. The concept of $x_{ave.}$ suppresses the computed energy density distribution for radial distances x close to the range of the most energetic delta ray, particularly at low ion velocity where delta rays of maximum energy are abundant.

The expressions for W and W_x can now be expressed as functions of x, from Eqs. (10) and (13), as

$$W(x) = w(2X - 2x) \tag{14}$$

and

$$W_x(x) = w(2x), \tag{15}$$

where X = 0.5r.

In the computation it is convenient to compile the information for $W\eta$ in functional form as

$$W\eta/W_x = f(w/W_x), \tag{16}$$

where

$$f(w/W_x) = \begin{cases} A' + (a + bw/W_x + cw^2/W_x^2)^{\frac{1}{2}}, \text{ for } w/W_x \leq u \\ (w^2/W_x^2 - d)^{\frac{1}{2}}, \text{ for } w/W_x \geq u. \end{cases} \tag{17}$$

The Eq. (17) fits the relationship defined by Eq. (16) to within 1% in the region $0.1 \leq r_o \leq 10.0\mu$, with the values of the constants being A' = -0.3296, a = 2.138, b = -4.058, c = 2.029, d = 2.545, and u = 2.920. Equation (16) is plotted in Plate II. When using this formulation for different materials and different values of r_o, the functional form of Eq. (17) remains the same but the constants A', a, b, c, d, and u are changed, as demanded by Eq. (16). At the appropriate energies and dimension in aluminum, Eq. (16) is in good agreement with the appropriate curve of Kanter, (reference 5, Fig. 10b).

By multiplying and dividing the integrand of Eq. (4) by W_x and then substituting for dn/dw from Eq. (5) and for $W\eta/W_x$ from Eq. (16), the energy density equation becomes

$$E^+ = [KZ'^2/(\beta^2 x)](\partial W_x/\partial x)(\partial\ /\partial W_x)\left(\int_{W_x}^{W_{max}} W_x f(w/W_x)(1/w^2)dw\right), \tag{18}$$

where $K = e^4 N/(mc'^2)$.

After substituting for $f(w/W_x)$ from Eq. (17) and carrying out the indicated integration and partial differentiation, the energy density can be expressed as

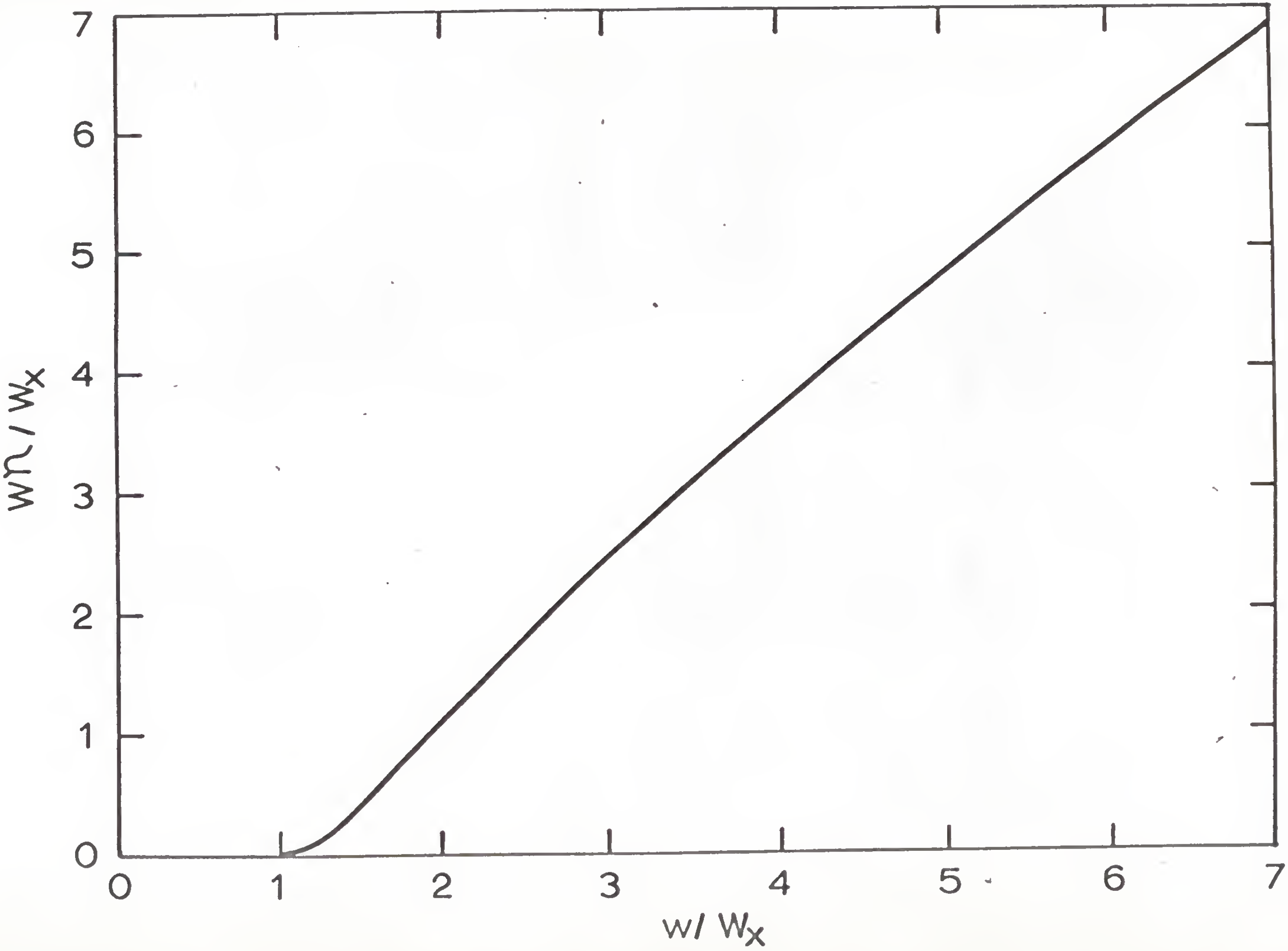
$$E^+ = \begin{cases} [KZ'^2 Q/(\beta^2 x)](\partial W_x/\partial x), \text{ for } W_{max}/W_x \leq u \\ [KZ'^2 Q'/(\beta^2 x)](\partial W_x/\partial x), \text{ for } W_{max}/W_x \geq u, \end{cases} \tag{19}$$

EXPLANATION OF PLATE II

Plot of $W\eta/W_x$ Versus w/W_x

for $0.1 \leq r_o \leq 10.0\mu$.

PLATE II



where

$$Q = -[A' + (aW_x^2 + bW_xW_{max} + cW_{max}^2)^{\frac{1}{2}}/W_x]/W_{max}, \tag{20}$$

$$Q' = -(W_{max}^2 - dW_x)^{\frac{1}{2}}/(W_xW_{max}), \tag{21}$$

and

$$\partial W_x/\partial x = \{C+[2C^2x+AC(B+1)]/[4C^2x^2+4AC(B+1)x+(AB-A)^2]^{\frac{1}{2}}\}/(AC). \tag{22}$$

Since Eq. (19) is only a function of x, β, N, ρ, and Z, one can calculate the density of ionization energy at a distance x from the path of a passing ion of atomic number Z and speed $\beta c'$ for a medium of density ρ and electron density N. Errors are introduced into the calculation by the fact that η has a slight dependence on the atomic number of the medium[6], by the assumption that the delta rays are totally diffuse, and by the use of the average "transverse distances", x. The computation then incorporates available data on electron penetration in matter in a manner compatible with the requirements of machine computation, which is a limitation on the solution.

TRACK WIDTH THEORY

Since the detection of heavy ion tracks in electron sensitive emulsion, in 1948, several methods have been proposed for identifying the ion causing the formation of the track. These methods are counting the number of delta rays per unit length of track, measuring the length of a delta ray, measuring the ejection angle of a delta ray with respect to the ion's path, and measuring the track width as a function of residual ion range, as measured back from the position of the resting ion.

In part, the present research was directed to investigate the use of track width (as a function of ion range) as a possible means of identifying the ion.

Several previous theoretical studies[7,8,9,10] of track width have been carried out by other investigators with only partial success. In these studies the criteria for computing the track boundary were the number of delta rays per unit length of track, energy flux, and energy density. In the investigations reported in this thesis, the criterion of energy density is used for defining the track boundary.

When passing through electron sensitive emulsion, and ion and its associated delta rays lose their energy by ionizing the atoms of the emulsion. According to Hamilton and Bayer[11], the initial mechanism for the formulation of a latent-image is the creation of electron-hole pairs. Specifying the creation of an electron-hole pair as a hit, an emulsion grain appears to be sensitized in a multi-hit (30 - 100 hits) process, so that the response is a rapidly changing function of ionization energy, which may be approximated as a step function for present purposes. Thus it is reasonable to assume that the activation of an emulsion grain occurs when there is a sufficient density of ionization energy deposited in a grain to create the required number of electron-hole pairs. Therefore, the track boundary is computed as the surface of a cylinder centered on the ion's path at which the density of ionization energy is a constant, E^*. According to this criterion, all the grains will be activated that lie within a cylinder of radius x centered on the ion's path, where at the distance x the density of ionization energy is E^*.

The track width, 2x, was calculated as a function of Z and β from Eq. (19) by setting $E^+ = E^*$, $N = 1.048 \times 10^{24}$ electrons/cm^3, and $\rho = 3.812$

grams/cm^3. These values for N and ρ are for G-5 emulsion.

To achieve agreement between experimental and computed track shapes, and to obtain a cosmic ray heavy ion spectrum similar to distributions reported by others[12], a value of 6500 ergs/cm^3 was chosen for E^*.

In order to obtain track width as a function of Z and ion range, R, Barkas'[13] range-β relation for heavy ions was used,

$$R(\beta) = (M/Z^2)[\lambda(\beta) + B_z(\beta)], \tag{23}$$

where

$$B_z = \begin{cases} 1.296\text{X}10^{-3}\beta Z^{5/3} \text{ cm, for } \beta \leq 2Z/137 \\ \\ 1.897\text{X}10^{-5}Z^{8/3} \text{ cm, for } \beta \geq 2Z/137, \end{cases} \tag{24}$$

M is the mass of the ion in units of a proton, and $\lambda(\beta)$ is the range of an ideal proton as a function of β. (An ideal proton is a particle of protonic mass and charge that does not capture electrons or interact strongly with nuclei.) The mass M was taken as the mass of the most abundant naturally occurring isotope, and the data for the range of an ideal proton was taken from Table 10.4.1 of Barkas[1].

The track width as a function of ion range and Z is plotted in Plate III.

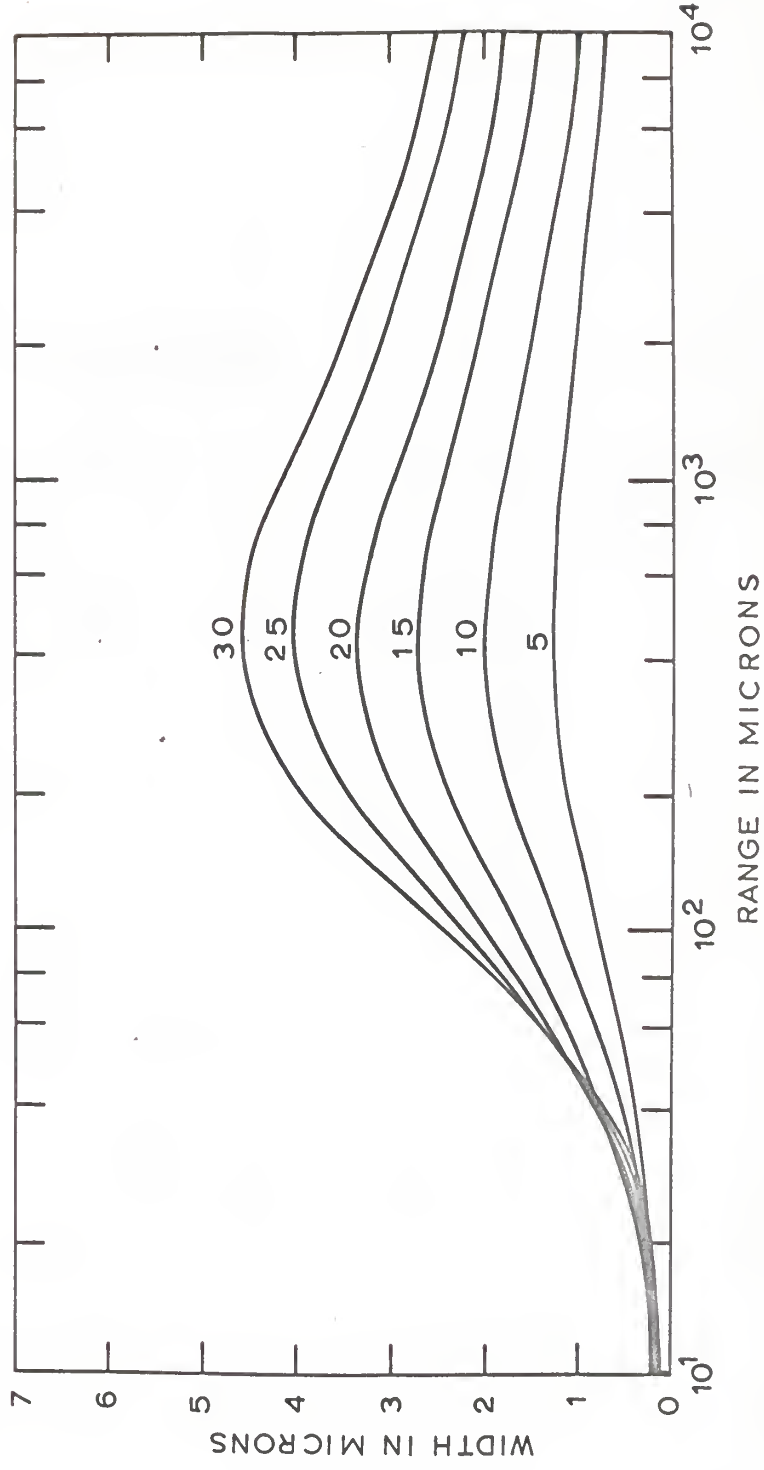
EXPERIMENT

Using a stereo-zoom microscope at 40X magnification, emulsion plates exposed in two balloon flights from Fort Churchill in July 1962 (and processed by D. E. Guss) and July 1965 (and processed by M. W. Friedlander), which were processed to the same developed grain diameter (approximately 0.8μ), were scanned for heavy ion tracks with a dip angle less than 10 degrees.

EXPLANATION OF PLATE III

Plot of Calculated Track Width Versus
Ion Range for $Z = 5, 10, \ldots, 30$.

PLATE III


WIDTH IN MICRONS
0
1
2
3
4
5
6
7
RANGE IN MICRONS
10^1
10^2
10^3
10^4
30
25
20
15
10
5

Photographs were then taken of the tracks at specified ranges by using a Leitz Ortholux microscope with an Aristophot attachment, a 100X plano oil immersion objective in conjunction with a 6X paraplan eyepiece, and a 70 mm, 100 ft. roll, camera back. From these photographs, 8"X10" glossy prints were made. The total magnification of the tracks is about 3380X.

The data from the 8"X10" prints were taken by tracing around the image of the tracks according to a specified criterion; that one grain of each delta ray track protruding from the ion track would be considered as part of the track image. Because of the roughness in the track boundary, the width was determined by measuring the area per unit length of a segment of track. The segment lengths used were 10μ in the first 100μ of track, 50μ between the ranges of 100 - 1000μ, and 100μ for greater ranges. These segment lengths were such that the average width did not change appreciably in the segment length. The areas were measured by use of a planimeter.

The tracks were traced by different individuals on different prints from the same negative using the same tracing criterion. The data used in this thesis were taken from 17 tracks, each traced by 4 individuals, 43 tracks, each traced by 3 individuals, and 16 tracks, each traced by 2 individuals. The agreement in tracing between the different individuals was about 5% for all 76 tracks.

To reduce the error introduced by differences in tracing between individuals and variations in emulsion processing, each tracing was normalized by subtracting from all width measurements the average width in the first 30μ of track.

The Z assignments were based entirely on the agreement between experiment and theory for the area between 150 - 300μ and the average width between 300 - 600μ. These are the regions of the track where there is best Z discrimination. In the region 150 - 300μ, the width changes most rapidly with range, and the track boundary is smooth. In the region 300 - 600μ, the width changes most rapidly with Z, and the average width is the maximum width of the track.

A plot of the average width from 300 - 600μ versus the area from 150 - 300μ for theory and experiment is given in Plate IV.

Additional work is being undertaken by M. R. Querry in this laboratory to examine the consistency of assignments by track width with Z assignments by delta ray counts. Results thus far obtained imply that the two mehtods are internally consistent.

Attempts at other internal checks of Z, such as the range and angular distribution of delta rays, were unsuccessful.

Other attempts to calibrate the Z spectrum by means of machine accelerated ions from the Berkeley accelerator yielded inconclusive results, possibly because machine accelerated ions have a range of less than 150μ, and are entirely in the top 15μ of emulsion, where the emulsion is quite sensitive to processing procedures. Cosmic ray ion tracks selected for study in this work generally begin at about 100μ from the surface of the emulsion.

RESULTS

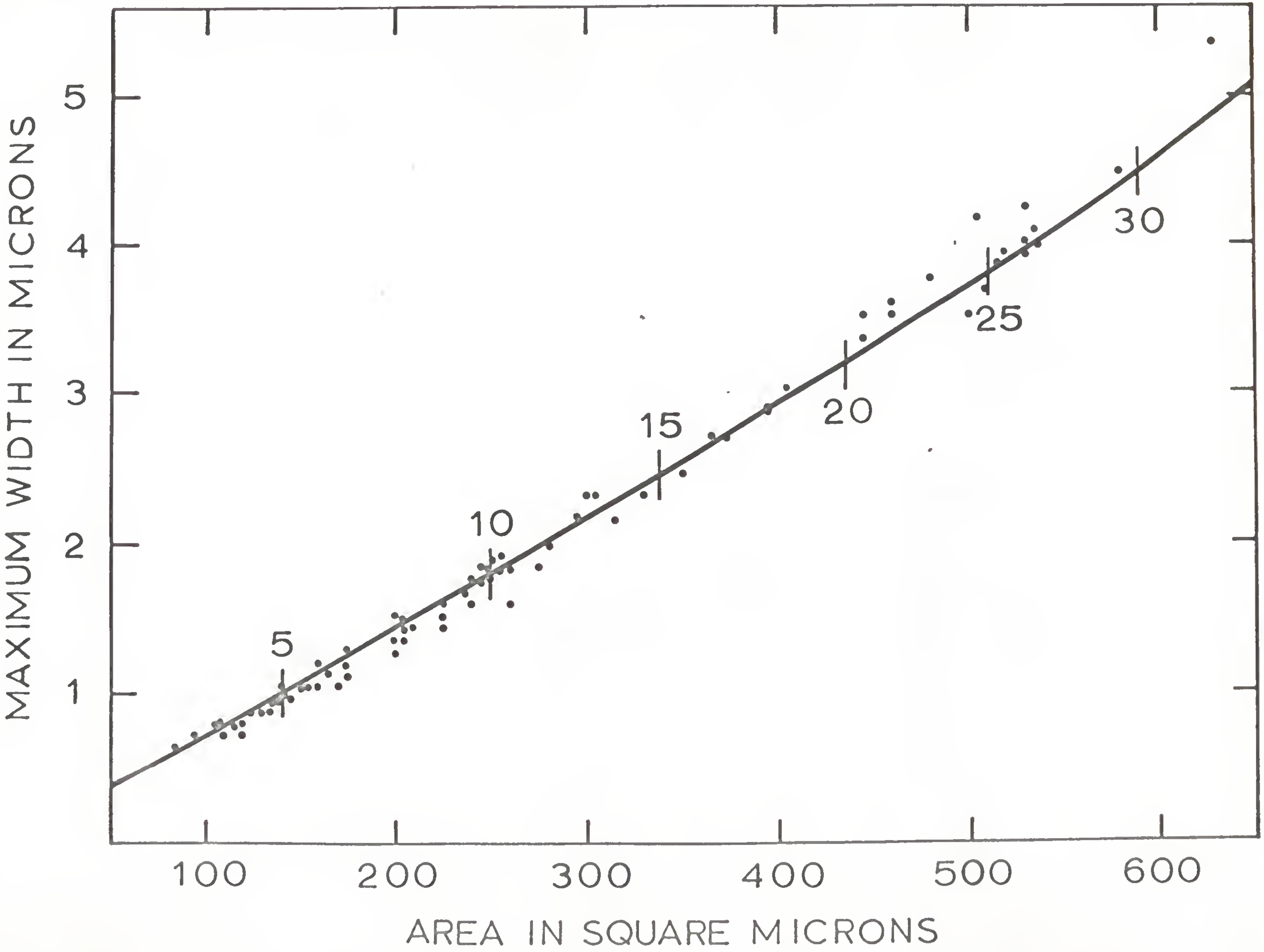
Due to the capture of electrons by the ion at low velocities, the theory predicts crossing over of the width-range curves below 50μ as shown in Plate III. This prediction of cross over is not detected experimentally because the magnitude of the cross over (0.1μ) is less than an undeveloped grain diameter (0.27μ).

In order to compare the normalized experimental data to theory, the

EXPLANATION OF PLATE IV

Plot of the Maximum Width (Average Width Between 300 - 600μ of Ion Range) Versus the Area (150 - 300μ of Ion Range) for 76 Experimental Tracks and Computation for Z = 1 - 35.

PLATE IV


MAXIMUM WIDTH IN MICRONS
AREA IN SQUARE MICRONS
5
4
3
2
1
100
200
300
400
500
600
5
10
15
20
25
30

average theoretical width for each Z in the region 0 - 30μ was subtracted from all calculated widths.

The Z assignments taken from Plate IV by averaging the width assignment and the area assignment for each of the 76 tracks used are listed in Table 1. To make the best comparison of experiment to theory, the width of the tracks of the same Z assignment were averaged (with the exception that the tracks assigned as Z = 26 and 27 were averaged together).

Table 1. Z assignments.

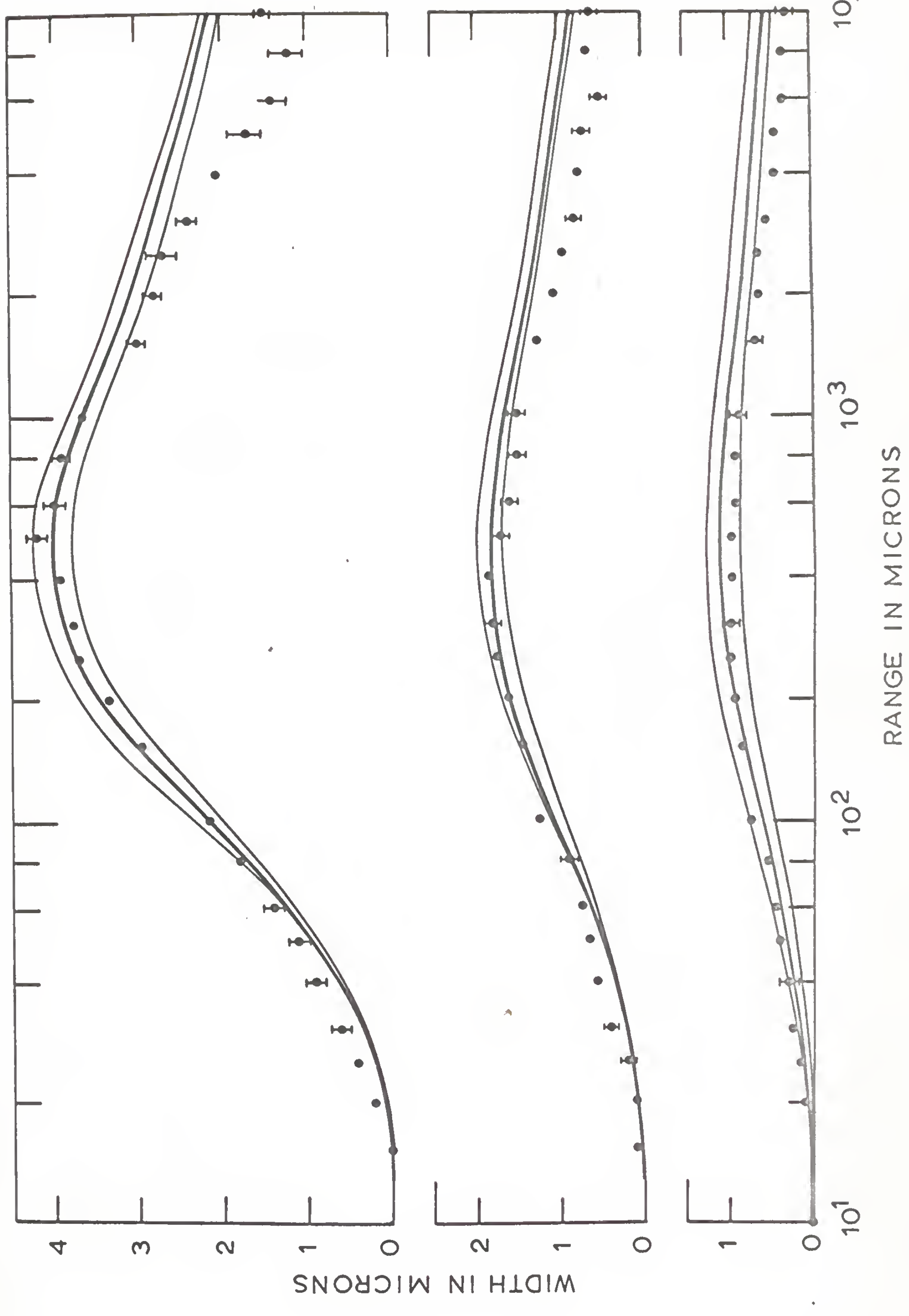
Z	No. of tracks	Z	No. of tracks	Z	No. of tracks	Z	No. of tracks
3	3	10	8	18	1	26	4
4	7	11	3	19	1	27	4
5	9	12	1	21	1	30	1
6	6	13	3	22	2	36	1
7	2	14	1	23	1		
8	8	15	1	24	3		
9	2	16	2	25	1		

In Plate V, the averaged width for all tracks assigned Z = 5 is plotted over the theory for Z = 5 $\pm$ 1 and similarly for Z = 10 $\pm$ 1 and Z = 26 $\pm$ 2. As shown in Plate V, there is good agreement between experiment and theory only in the region 100 - 700μ of ion range where the Z discrimination is less than $\pm$ 2. At ion ranges less than 100μ, the theoretical width is less than the experimental width. This disagreement is due to the concept of the average transverse distance, $x_{ave.}$, which does not allow a delta ray to deposit energy at a distance greater than one half of its true range from the ion's path. There is, however, a finite probability that a delta ray may even deposit energy at a distance equal to its true range from the ion's path. This probability plays a more important role at ion ranges less than

EXPLANATION OF PLATE V

Plot of the Averaged Experimental Track Width for Tracks Assigned $Z = 5$, 10, and 26-27 Versus Range Over Computed Track Width for $Z = 5 \pm 1$, $Z = 10 \pm 1$, and $Z = 26 \pm 2$.

PLATE V



100μ than at greater ranges because of the severalfold increase in the number of delta rays that receive energy W_{max}. In the present work, this probability function is not considered because the form of the function is not known, because of machine limitations, and because this region of the track contains little Z discrimination.

At ion ranges greater than 700μ, the theory predicts widths that are greater than experiment. This disagreement is not completely understood. We do know, however, that the agreement is not improved by using the relativistic delta ray distribution or different angular distributions. There is reason to suspect that part of the disagreement is in the range-β relation for heavy ions. The range-β relation for an ideal proton used in this work as given by Barkas[1] in 1963 (which is in agreement with Shapiro[14]) disagrees by about 20 - 30% with a subsequent range-β relation again given by Barkas[13] in 1964. The experimental work of Rudd, Sautter, and Bailey[15] implies that the $1/w^2$ term in Eq. (5) varies from $1/w^{1.8}$ to $1/w^{2.28}$ for low Z and at low ion energies in gases of low atomic number, while Gryzinski[16] computes a $1/w^3$ term. Also the experimental results of delta ray counting by Aizu[17] and his co-workers for heavy ions in emulsion state that the number of delta rays varies as Z^2 in the region about 1000μ in ion range and as $Z^{1.5}$ at greater ranges. These questions are in need of further experimental test. Error is also introduced because we are dealing with bound electrons, while the delta ray distribution formula, Eq. (5), used is strictly valid only for free electrons.

CONCLUSIONS

Some of the reaining difficulties in the calculation are the concept of the average transverse distance, the uncertainties in the range-β relation for heavy ions, and the uncertainties in the delta ray distribution function. With the availibility of larger and faster computers, the problem of the average transverse distance can be corrected. Emulsion studies provide the only experimental access presently available to studies of the delta ray distribution function at high energies. Disagreements between calculated and measured track width at high energies imply that research in this field is desirable.

The stronger points in the calculation of the spatial distribution of ionization energy are the incorporation of the latest electron penetration data into the computation and the versatility of the formulation to be used for calculations in different materials and different dimensions. Track profiles developed from these calculations are in excellent agreement with observed tracks between the ion ranges of 100μ and 700μ leading to a superior system of Z discrimination.

ACKNOWLEDGEMENTS

Appreciation is expressed to Dr. Robert Katz for his helpful suggestions and criticisms, to Marvin Querry for his helpful suggestions and his help in obtaining the experimental data, to D. E. Guss and M. W. Friedlander for emulsion flights and processing, to H. J. Goldberg for processing and tracing, to C. R. Criss for his excellent photographic printing, to the 1410 Computer Center for the use of the machine, and to Dorothy Koepsel, K. G. Shah, Makadam Saleh, Qassim Tabatabai, Max VanGassbeek, Abdulilah Kanawi, and George Hofer for their help in tracing and making measurements.

Appreciation is also expressed to my wife, Gloria, who typed this thesis.

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B. S., Kansas State University, 1965

AN ABSTRACT OF A MASTER'S THESES

submitted in partial fulfillment of the

requirements for the degree

MASTER OF SCIENCE

Department of Physics

KANSAS STATE UNIVERSITY
Manhattan, Kansas

1967

Due to the increasing interest in radiation damage by heavy ions, its hazards and its applications, an accurate calculation of the spatial distribution of ionization energy associated with a heavy ion is needed. This distribution has been calculated using the latest electron penetration data and a highly versatile formulation that can be used for different media and different spatial dimensions.

The calculated energy distribution, when applied to the width of ion tracks in emulsion, gives good agreement with measurements obtained by tracing around the photographic image of a track that has been magnified 3380X. Using the same tracing criterion, each track was traced by two or more individuals whose tracings agreed to within about 5%. The Z assignments made from the area between the ion ranges of 150 - 300μ and the maximum width of the track (the average width between the ranges of 300 - 600μ) by agreement between experiment and theory are in good agreement with Z assignments based on delta ray counts.

The disagreement between calculated track width and experiment at ion ranges greater than 1000μ, may lead to a better formulation of the delta ray distribution generated by rapidly moving heavy ions.