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IONIZATION FLUCTUATIONS IN CELLS AND THIN DOSIMETERS

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Berkeley, California

UNIVERSITY OF CALIFORNIA

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Berkeley, California

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ERRATUM

TO: All recipients of UCRL-17465

FROM: Technical Information Division

Subject: UCRL-17465, "Ionization Fluctuations in Cells and Thin Dosimeters," H. D. Maccabee and M. R. Raju, March 15, 1967.

Please make the following correction on subject report.

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ABSTRACT

Since fast charged particles lose energy in matter by a collision process which is discrete and random, statistical fluctuations are expected in the energy loss of such particles when traversing "thin" absorbers. The theory of ionization fluctuations has been developed by Bohr, Landau, Symon, Vavilov and others, and has been verified by several experimenters, including Maccabee and Raju. Cells and thin dosimeters acts as thin absorbers for many types of particulate radiation, and thus significant fluctuations in energy deposition are to be expected. We discuss the application of the theory to these cases, and the effect of energy-loss straggling on the Bragg peak of charged particle beams.

I. INTRODUCTION

When energetic charged particles pass through matter, they lose energy predominantly by a series of inelastic collisions with the electrons of the material, resulting in ionization and excitation of the atoms of the material. Since the collisions are discrete and random, statistical fluctuations in ionization are expected.

In first approximation, the probability of energy loss ϵ in a single electronic collision is proportional to ϵ^{-2}. If this collision spectrum is summed over all possible collision energy losses, we obtain an expression for the average linear rate of energy loss due to ionization and excitation. The standard formula for this quantity (for particles heavier than electrons*) is:

$$\frac{dE}{dx} = - \frac{4\pi e^4 z^2 NZ}{mv^2} \left[\ln \frac{2mv^2}{I(1-\beta^2)} - \beta^2 - \frac{C}{Z} - \frac{\delta}{2}\right], \qquad (1)$$

*Note on electrons: although many of the arguments presented here are valid for electrons, we consider only heavy charged particles in the following treatment.

where
e = electron charge,
z = particle charge number,
N = number of atoms per cm^3 of material,
Z = atomic number of material,
m = electron mass,
v = particle velocity,
I = mean excitation potential of material ≈ 13.5 Z(eV),
β = particle velocity ÷ speed of light,
$\frac{C}{Z}$ = shell correction (negligible for protons > 1 MeV),
$\frac{\delta}{Z}$ = density correction (negligible for protons < 1 GeV).

This quantity is often called the stopping power S; if all the energy lost is "imparted locally" to the medium, then S is identical with L, the linear energy transfer (often denoted by LET). The product of the linear energy transfer and the particle fluence Φ (the number of particles entering per unit area) yields the energy imparted per unit volume, which may be multiplied by the density to give the absorbed dose.

In a thin slab of matter (one in which the energy loss is small compared with the total kinetic energy of the particle) we can assume that the average energy loss rate is approximately constant through the slab, and thus write for the average total energy loss $\overline{\Delta}$ in thickness x:

$$\overline{\Delta} = \left(\frac{dE}{dx}\right)(x). \qquad (2)$$

The ϵ^{-2} dependence of the collision spectrum implies that collisions resulting in a large energy transfer to an electron are relatively rare compared with small-energy-transfer collisions. Although they are relatively infrequent, the large-energy-transfer collisions account for a significant proportion of the total energy loss. The relatively high energy electrons resulting from these rare collisions are often called delta rays. In a thin absorber, the probable number of large-energy-transfer collisions may be so small that the random statistical variations in this number are relatively large, and result in significant fluctuations in the energy lost in this mode; thus fluctuations occur about the average total energy loss, $\overline{\Delta}$. These fluctuations are often called energy-loss straggling.

II. THEORY

Since the fluctuations depend on the number of large-energy-loss collisions, a dimensionless parameter which provides an estimate of this number should be useful to characterize the distribution of total energy losses. Such a parameter, κ (kappa), was introduced by Vavilov in his exact theoretical treatment of ionization fluctuations.[1]

$$\kappa \equiv 0.150 \left(\frac{sZz^2}{A}\right)\left(\frac{1-\beta^2}{\beta^4}\right), \qquad (3)$$

where
s = thickness of absorber in $g/cm^2 = \rho x$,
A = atomic weight of absorber,
Z, z, and β are as defined above.

As the absorber thickness increases and the particle velocity decreases, κ increases, corresponding to the increased number of particle-electron collisions in the highest collision-energy interval. The case of $\kappa \gg 1$ was treated in 1915 by Bohr,[2] who found that the distribution of total energy losses is Gaussian, with variance given by

$$\sigma^2 = 0.157\ sZz^2/A \quad [\text{in}\,(\text{MeV})^2], \qquad (4)$$

and the most probable energy loss equals the mean.

For thinner absorbers and higher particle velocities, κ decreases and fluctuations become much more severe. The case of $\kappa \leq 0.01$ was treated in 1944 by Landau, who found a broad asymmetric distribution characterized by a long high-energy-loss "tail" and a most probable energy loss which is considerably less than the average.[3] The full width of the Landau distribution at half maximum is given by

$$\text{FWHM} = 0.611\ sZz^2/A\,\beta^2\ (\text{in MeV}), \qquad (5)$$

and the most probable energy loss is

$$\Delta_{mp} = \frac{2\pi e^4 z^2 NZ}{mv^2} \times \left[\ln \frac{4\pi e^4 NZ}{I^2(1-\beta^2)} - \beta^2 + 0.37\right]. \qquad (6)$$

There are many cases corresponding to intermediate values of κ, i.e., $0.01 \leq \kappa \leq 1$; these cases were treated approximately by Symon[4] in 1948, and exactly by Vavilov[1] in 1957. See Fig. 1. As might be expected, the energy-loss distributions for these cases form a smooth transition between the narrow symmetric Gaussian and the broad highly-skewed Landau distribution. The Vavilov theory is general, and includes the Gaussian and Landau distribution as its special cases. The numerical quadrature of Vavilov's rigorous but complicated solution was performed by Seltzer and Berger[5] in 1964. They provide a systematic and comprehensive tabulation of the Vavilov distribution in terms of the parameters κ and β^2, and furnish tables relating κ and β^2 to the absorber thickness and particle energy.

III. EXPERIMENT

There is extensive experimental evidence for the validity of the Bohr and Landau theory of energy-loss fluctuations, but until recently there have been few data in confirmation of the more general Vavilov formulation. Maccabee and Raju have used solid-state silicon semiconductor detectors to measure the energy-loss distributions of protons up to 730 MeV and alpha particles up to 910 MeV in order to verify the quantitative theory of ionization fluctuations over virtually the whole range of the significant parameter κ (Ref. 6). Semiconductor detectors have several advantages for this type of measurement: their density (and thus their stopping power) is about a thousand times that of a gas, yielding that many more energy-loss collisions per unit path length. Also the energy required to create a charge pair in silicon is 3.6 eV (approximately a tenth of the value for gas), yielding ten times as many charge pairs. The result of these properties is to improve the charge statistics and thus yield superior energy resolution. In addition, the semiconductor detectors have relatively uniform sensitive thicknesses, highly linear response, and short pulse duration. The method of the experiment is to pass a parallel monoenergetic beam of heavy charged particles (from an accelerator) through the detector and measure the pulse-height spectrum with a multichannel analyzer. The system is calibrated by using standard sources.[6] Since the pulse height is directly proportional to the energy loss in the detector, the pulse-height distribution may be simply processed to yield the energy-loss distribution, i.e., a plot of relative probability versus energy loss.

The results of a few of these experiments follow. Figure 2 shows the energy-loss distribution of 45.3-MeV protons in 0.265 g/cm^2 silicon (about 1mm) with $\kappa = 2.23$. The distribution is very close to a symmetric Gaussian with the most probable energy loss only 0.7% less than the mean energy loss, and an rms deviation (σ) of 145 keV, in agreement with the Bohr theoretical prediction.

Figure 3 shows the energy-loss distribution of 910-MeV alpha particles (He^{2+} ions) in 0.206 g/cm^2 silicon, with $\kappa = 0.318$. This curve is a good example of the intermediate values of κ, in which the distribution is asymmetric, with the beginnings of a high-energy-loss tail, and a most probable energy loss which is significantly (6%) less than the mean. For this curve, the value of the full width at half maximum is 22% of the mean energy loss, in agreement with the prediction of the Vavilov theory.

Figure 4 shows the energy-loss distribution resulting from 730-MeV protons passing through 0.413 g/cm^2 silicon, with $\kappa = 0.021$. This is a good example of the lower range of κ, where the Landau theory is valid: the curve is highly asymmetric, with a long high-energy-loss tail, and a most probable energy loss which is 18% less than the mean. The full width at half-maximum is 180 keV, in agreement with the Landau theory. In general, there is very good agreement between the measured experimental energy-loss distributions and the Vavilov theoretical predictions over virtually the whole significant range of κ (from $\kappa = 2.23$ to $\kappa = 0.003$).

Measurements of this type have been performed in gas detectors by several groups. Gooding and Eisberg[7] found good agreement with the Symon theory for 37-MeV protons in 1957, and Rosenzweig and Rossi[8] did a detailed study of energy-loss straggling for 5.8-MeV alpha particles in a variable-thickness proportional counter in 1963. They found general agreement with the Symon theory for κ values from 0.11 to 3.56, provided that corrections were applied for the effects of electron binding and delta-ray escape from their detector. Glass and Samsky have found agreement with the Vavilov theory of ionization fluctuations for protons of energy as low as 1 MeV in a gas detector equivalent to 0.5 micron of tissue. These results imply that the theory of ionization fluctuations can be applied to absorbers as small as cells and their constituents.

There is a limitation on the Bohr-Landau-Vavilov theory of ionization fluctuation, however. The theory is formulated in terms of continuum statistics, and thus depends on a large number of collisions occurring in at least the lowest collision-energy interval. Thus if the absorber is so thin that the mean energy loss is not much greater (say a factor of 20) than the mean excitation potential, there are so few collisions altogether that continuum statistics are invalid, and discrete Poisson statistics must be used. Examples of measurements for solids in this energy-loss region are given by Rauth and Simpson[10] for 20-keV electrons and Morsell for 992-keV protons.[11]

IV. APPLICATIONS

It is generally accepted that one of the most important parameters for characterizing radiation effects is the absorbed dose. As shown above, there are wide fluctuations in the energy loss for many cases of charged particles passing through thin absorbers, and thus we can expect fluctuations in the dose delivered by each individual particle. The result of this phenomenon can perhaps best be understood by considering another important parameter of radiation effects, the local energy deposition. The effect of energy-loss straggling is that, even for monoenergetic incident particles, the local energy transfer is not single-valued, but spread over a spectrum. The theory of ionization fluctuations can be used to predict the energy transfer spectrum, if corrections are made for the energy that is not imparted locally. In fact, measurements of the type shown above are actually measurements of the energy transfer spectrum in the detector.

There is a factor that mitigates the effect of energy-loss straggling to some extent: The largest fluctuations are due to the few highest energy collisions, which are just the collisions that produce the delta-rays that are most likely to escape the volume in question. The net effect is to transfer events from the high-energy-loss tail of the spectrum to the low-energy-loss end. One way of estimating the effect of delta-ray escape is by computing the "restricted" stopping power, i.e., the average energy-loss rate due to all collisions whose energy is less than that of a secondary electron that can just escape the volume in question. The approximate formula for the restricted stopping power was given by Bethe:[12]

$$\left.\frac{dE}{dx}\right|_{\epsilon<\epsilon_\delta} = -\frac{2\pi e^4 z^2 NZ}{mv^2}\left[\ln\frac{2mv^2\epsilon_\delta}{I^2(1-\beta^2)} - \beta^2\right], \qquad (7)$$

where ϵ_δ is the energy of a delta-ray electron whose range is equal to the dimension of the specimen, and it is assumed that $\epsilon_\delta << (2mv^2)/(1-\beta^2)$.

It is clear that biological cells act as "thin" absorbers for most forms of particulate radiation, and that significant energy-loss fluctuations will occur in many cases. For example, consider 10-MeV protons traversing slab-like cells of 5-micron thickness. In this case dE/dx is 4.7 keV/micron, and thus the mean energy loss in the cell will be about 24 keV. With $\rho \approx 1$ g/cm^3 and $Z/A \approx 0.5$, parameter $\kappa = 0.150\ s\ (Z/A)\ z^2(1-\beta^2/\beta^4) \approx 0.15\ (5\times10^{-4})\ 0.5(0.98/4.4\times10^{-4}) \approx 0.08$. Thus the Vavilov distribution holds, and the most probable energy loss in the cell will be only about 82% of the mean, and the full width of the energy-loss distribution at half maximum will be 8 keV.

Of course most cells are not slab shaped and most incident radiations are neither monoenergetic nor parallel. Thus in order to estimate the true distribution of energy deposition in the cell, the energy-loss distribution must be "folded in" with the path-length distribution in the cell and the effects of the distribution of energies in the incident radiation.

Several experimental methods have been developed to measure the parameters of dose quantity and quality in masses comparable to that of the cell. Notable among these thin dosimeters are the tissue-equivalent ionization chambers and proportional counters in slab and cylindrical geometry, and the system of spherical microdosimeters developed by Rossi and his colleagues.[13] Even a cursory examination of the results of such experiments is sufficient to show that ionization fluctuation is one of the primary factors determining the shape of the measured distributions, and that fluctuation theory should be applied in the analysis of the data. It should be remembered, however, that relative biological effectiveness is probably only a slowly varying (e.g., logarithmic) function of specific ionization, and thus even fluctuations of energy loss by a factor of 3 about the mean should not make a large difference in the results of a biological exposure. If an energy threshold exists for a biological effect, however, the effect of fluctuations should be more severe.

Naturally, if there are fluctuations of the energy lost in a given small thickness, one expects fluctuations in the total thickness traversed by particles in losing all their energy. This phenomenon is called range straggling and is the cumulative effect of energy-loss straggling over a large thickness of absorber. Since there are a large number of collisions at all energies, the range distribution is approximately Gaussian:

$$P(R) \approx \frac{1}{(2\pi)^{1/2}\sigma_R} \exp[-(R-\bar{R})^2/2\sigma_R{}^2], \tag{8}$$

where P(R) is the probability of range R, $\bar{R}$ is the mean range, and $\sigma_R{}^2$ is the variance. Berger and Seltzer[14] give a more thorough discussion of this subject and multiple scattering, and tabulate σ_R, which varies between 1 and 2% of the mean range for protons of 300 to 2 MeV in light elements. The net effect of range straggling and multiple scattering on a monoenergetic charged particle beam is to broaden considerably the peak of the Bragg ionization curve. The physical explanation of this effect is that some of the particles are stopping (and therefore ionizing heavily) while others still have enough kinetic energy to travel farther. The consequences of this effect can be clearly seen in Fig. 5, which shows the energy distribution of a 910-MeV alpha particle beam at the Bragg peak, and the corresponding LET values, as measured by Raju.[15] Note that the modal energy at the peak is much higher than one might expect. This case, along with our measurements on a 50-MeV proton beam and measurements at the Harvard cyclotron, indicate that one can use a general rule of thumb that the most probable energy at the Bragg peak is about 10% of the initial kinetic energy.

ACKNOWLEDGMENTS

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FIGURE CAPTIONS

Fig. 1. Normalized probability $\phi(\lambda)$ versus Landau's energy-loss parameter λ, for $\beta^2 = 0.5$. Note the smooth transition between the energy-loss distributions as parameter κ decreases from 10 to 0.

Fig. 2. Energy-loss distribution of 45.3-MeV protons in 0.265 g/cm^2 silicon; $\kappa = 2.23$.

Fig. 3. Energy-loss distribution of 910-MeV alpha particles in 0.206 g/cm^2 silicon; $\kappa = 0.318$.

Fig. 4. Energy-loss distribution of 730-MeV protons in 0.413 g/cm^2 silicon; $\kappa = 0.021$.

Fig. 5. Energy distribution measured at the Bragg peak of a 910-MeV alpha particle beam. LET values computed for water.

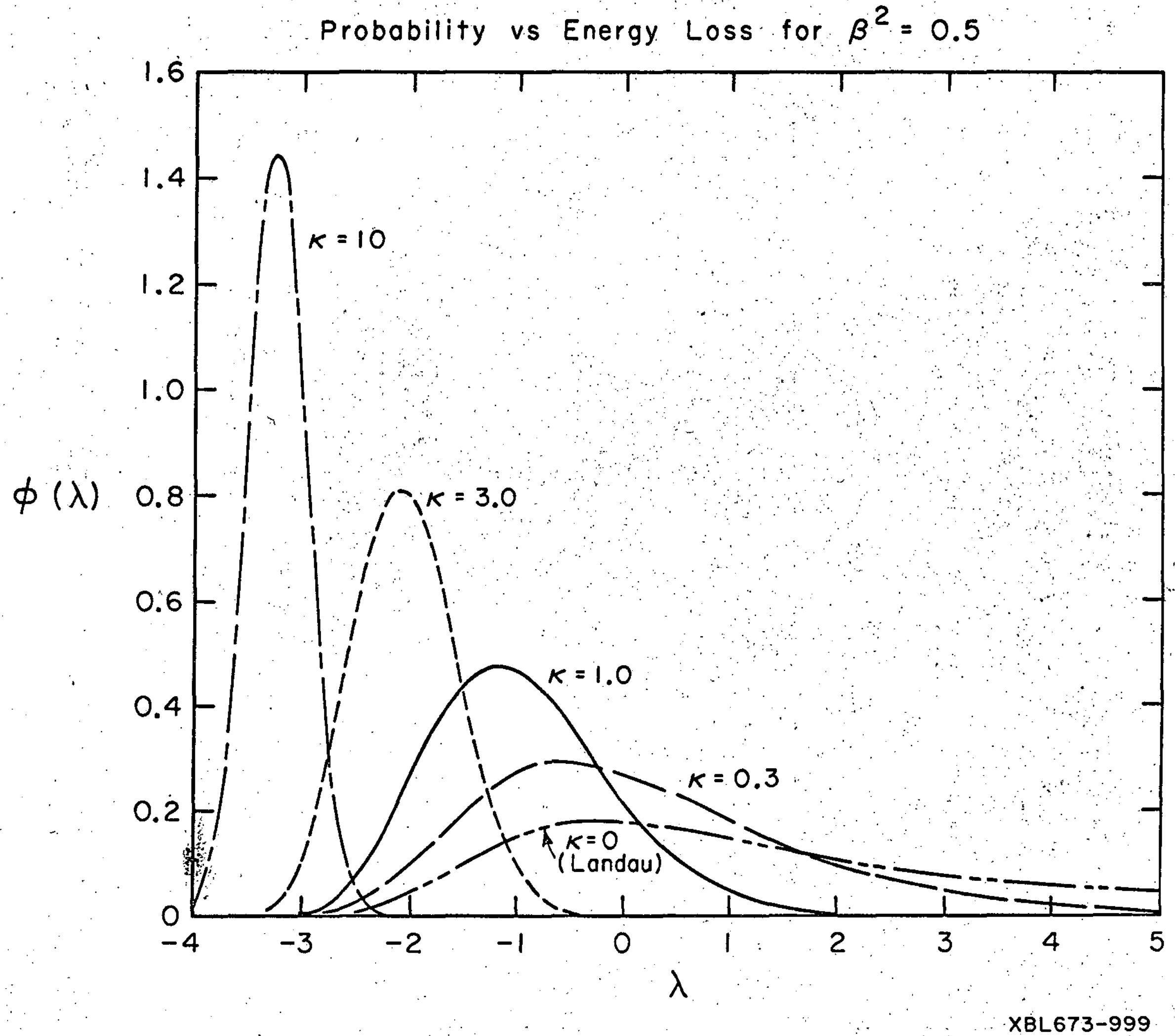

Probability vs Energy Loss for β^2 = 0.5
1.6
1.4
1.2
1.0
0.8
0.6
0.4
0.2
0
$\phi(\lambda)$
−4
−3
−2
−1
0
1
2
3
4
5
λ
κ = 10
κ = 3.0
κ = 1.0
κ = 0.3
κ = 0
(Landau)
XBL673-999

Fig. 1

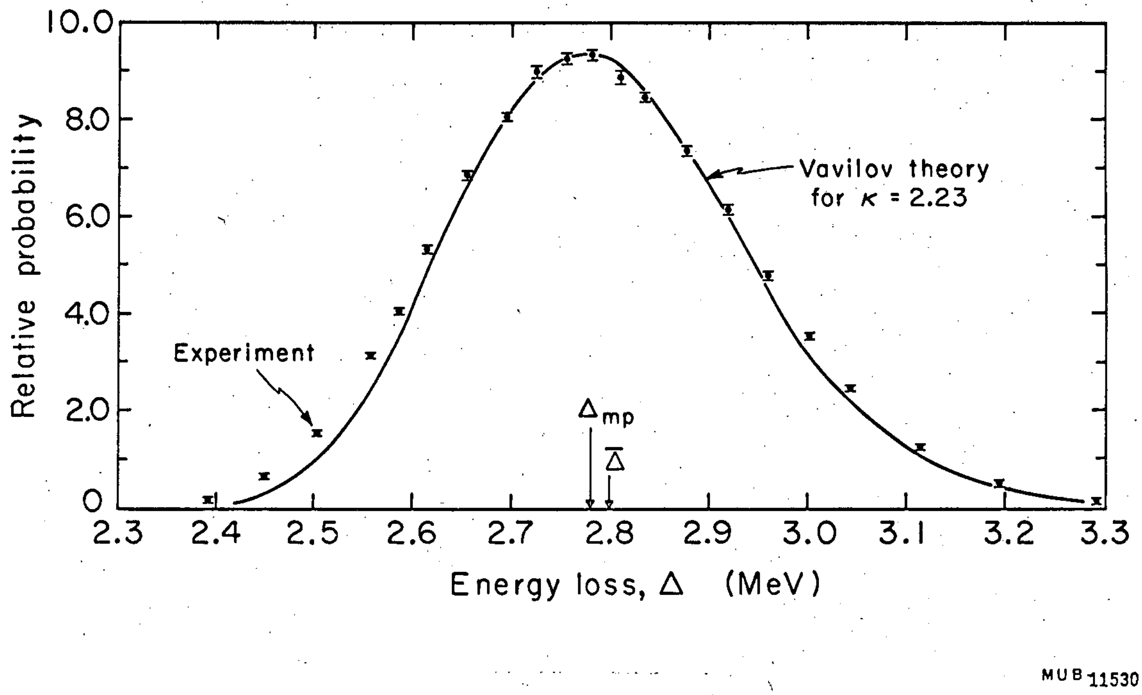
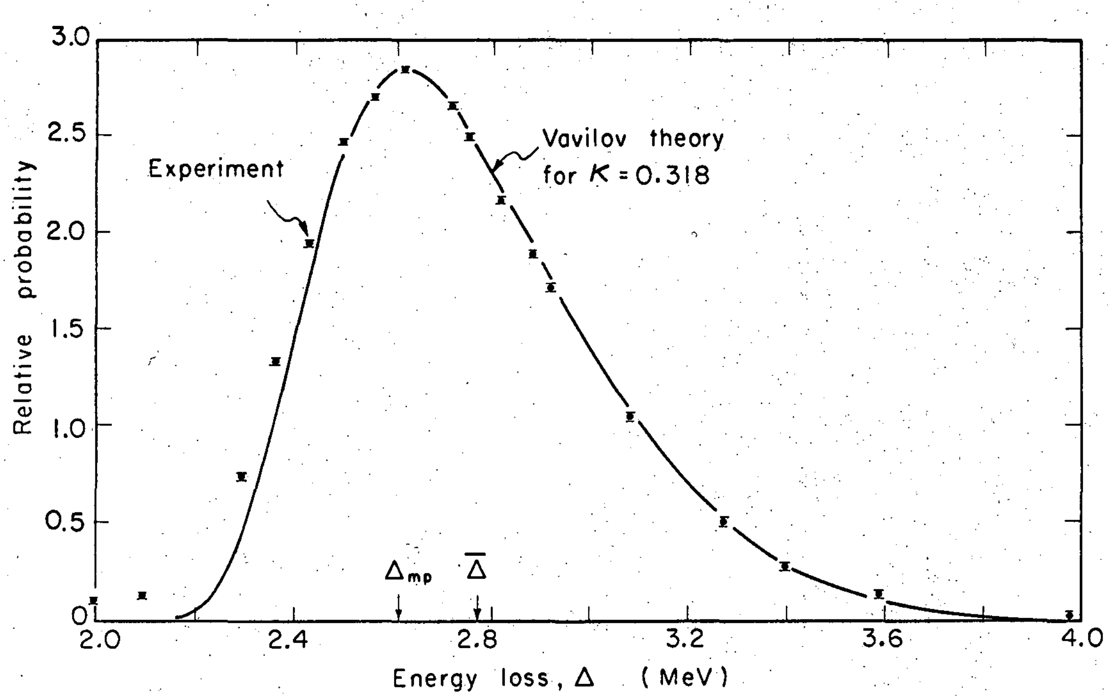
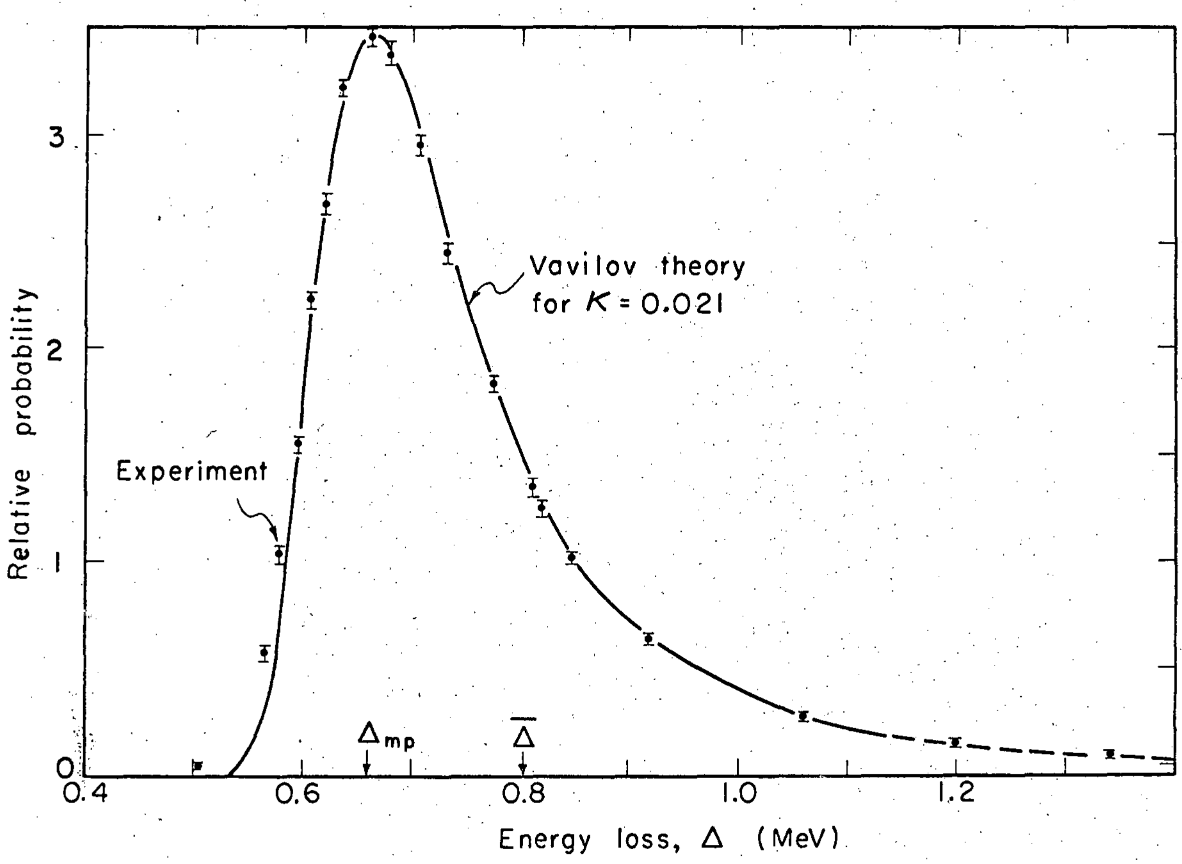

10.0
8.0
6.0
4.0
2.0
0
Relative probability
2.3
2.4
2.5
2.6
2.7
2.8
2.9
3.0
3.1
3.2
3.3
Energy loss, Δ (MeV)
Experiment
Vavilov theory
for κ = 2.23
Δmp
Δ̄
MUB 11530

Fig. 2



MUB 11434

Fig. 3



MUB-11432

Fig. 4

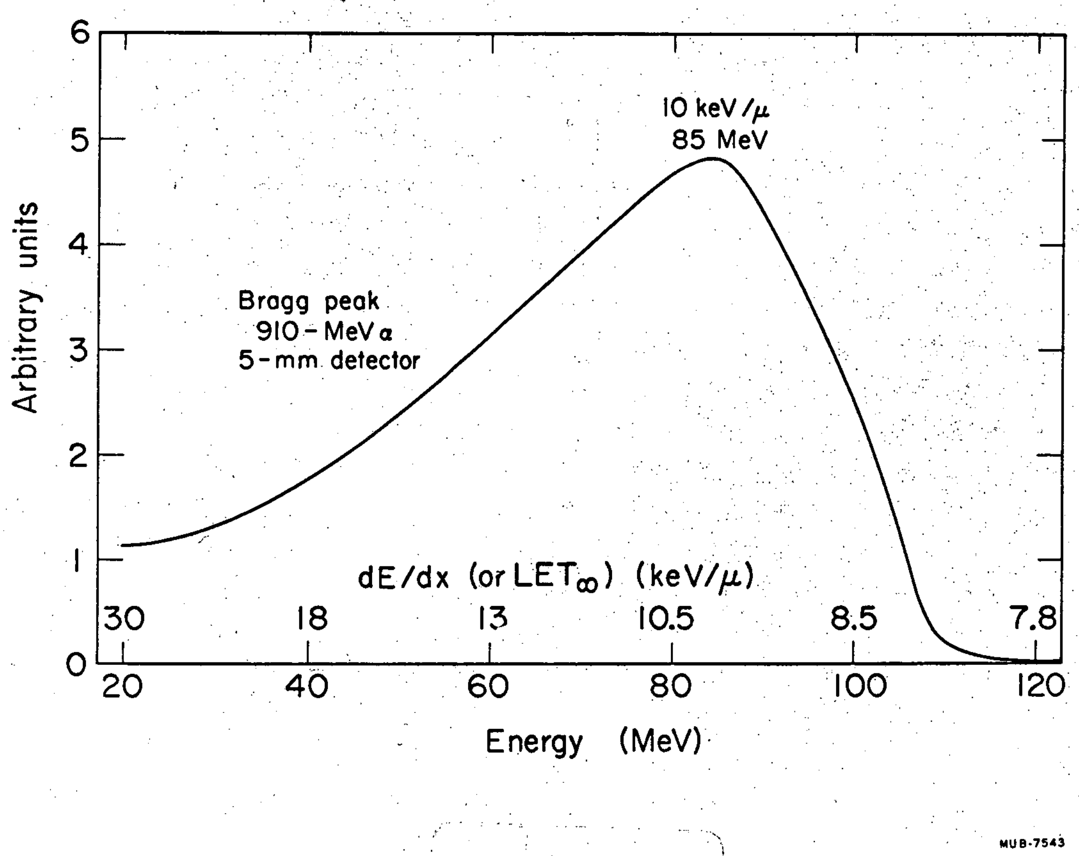

10 keV/μ
85 MeV
Bragg peak
910 - MeV α
5-mm detector
Arbitrary units
dE/dx (or LET∞) (keV/μ)
30
18
13
10.5
8.5
7.8
Energy (MeV)
MUB-7543

Fig. 5