Charged Particle Tracks in Solids and Liquids

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Contents

Parameters of track structure

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Abstract. Linear energy transfer (or collision stopping power) and energy straggling along the tracks of charged particles are both relevant to the effectiveness of ionizing radiation. Energy straggling is the dominant aspect whenever one is concerned with small energy depositions (<1 keV) and a correction to LET is necessary in these cases. The correction term and its relation to the spectrum of energy transfers in primary collisions is derived. Other parameters of track structure are discussed, and an analysis is mentioned which can substitute the Landau- and Vavilov-theory in the analysis of the collision spectrum.

1. Introduction

Track structure is an aspect of radiation quality disregarded in conventional LET theory but indispensable for an understanding of radiation effects. D. Lea's and L. H. Gray's pioneering works have made this clear. These works have also shown that the random patterns of primary events are a fairly complex result of different stochastic factors. A limited number of parameters cannot possibly represent the full picture.

A complete description involves the spatial patterns of all different quantum-mechanical interactions of radiation and irradiated medium. At present such a description is out of the question, and it is therefore necessary to simplify the problem. A crude but practicable approximation is to assume that the distribution of energy deposition is the same as for the spatial distribution of primary events and their reaction products. This assumption is, of course, untenable on a molecular scale up to distances of about 100 Å. Over a larger scale, however, the spatial patterns of energy deposition are an important and characteristic index of radiation quality. Experimental determination and theoretical analysis of these microscopic patterns are the essential points of Rossi's concept of microdosimetry (Rossi 1959, 1967).

Microdosimetry is a synthesis of LET theory and straggling. But the cases of greatest biological interest are those where one deals with energy depositions of less than a few keV; and in those cases straggling, i.e. the fluctuations of energy loss along the track of an ionizing particle, is the dominating factor (Kellerer 1968b). This is why the theory of microdosimetry is by and large a theory of track structure.

While explicit analysis is rather involved (Kellerer 1967, 1968a) some aspects of track structure can be discussed in a simplified way. This is done in the first part of this paper. Specifically it will be shown that the LET concept can be modified in such a way that it takes into account the 'clustering' of energy deposition along the track. In a second part of this paper different parameters of track structure are compared and it is shown how straggling experiments can be used to obtain information on these parameters and on the delta-ray spectrum.

2. LET and linear energy concentration

The effectiveness of primary events and of their reaction products depends on their local concentrations. Due to the discontinuous nature of energy deposition the term 'local concentration' must refer to track intervals of finite extension. The relevant length of these intervals is determined by the range of interaction of ionizations or radiation-induced free radicals. It may also be related to the size of sensitive structures in the cell. The assumption of a critical distance is reflected in the choice of an energy cut-off in LET theory and in the use of the corresponding 'restricted' LET values.

The reaction probabilities of the irradiation products in the trail of an ionizing particle depend on the whole spectrum of their mutual distances. In other words, one must know the probability distribution of local energy concentration around the activated molecules. But whenever a single index of radiation quality is necessary, one has to substitute the probability distribution by its mean value. This mean value will be derived here, and it will be seen that LET in its present form is not the relevant parameter.

When an ionizing particle of linear energy transfer L traverses a certain distance d in tissue it loses an energy E. The local concentration of energy over the interval can then be measured by the 'linear energy concentration' E/d. This quantity is a random variable; while its expected value is equal to L, its actual values deviate considerably from L. In order to indicate the somewhat loose relation to L, the linear energy concentration will in the following be designated* by the Greek letter Λ.

Let the discussion be limited to intervals which are small enough that the LET of the incoming particle does not change appreciably over the interval. The track segment can then be assumed to be a straight line. Under these conditions the probability distributions of E or Λ can be calculated. In fact this is the object of the classical 'thin foil' straggling theory of Landau (1944), Simon (1948) and Vavilov (1957) and of a recent more general solution of the straggling problem (Kellerer 1968a). An analysis of this problem and its implications in a wider context is found in Fano's paper of 1953. We will, for the moment, not be concerned with the explicit distributions, but only with some of their basic properties.

The probability distribution of energy transfer E can be designated by $f(E; n)$ the parameter n is the expected number of collisions along the interval d. The fraction of absorbed energy associated with values of Λ between E/d and $(E+\mathrm{d}E)/d$ is then equal to:

$$E\ f(E; n)\ \mathrm{d}E / \int E\ f(E; n)\ \mathrm{d}E.$$

In order to derive the value of Λ averaged over all elements $\mathrm{d}E$ of absorbed energy, one has to evaluate the integral:

$$\overline{\Lambda}_\mathrm{D} = \frac{\int \Lambda\ E\ f(E; n)\ \mathrm{d}E}{\int E\ f(E; n)\ \mathrm{d}E} = \frac{1}{d}\frac{\int E^2 f(E; n)\ \mathrm{d}E}{\int E f(E; n)\ \mathrm{d}E} = \frac{1}{d}\frac{\overline{E^2}}{\overline{E}}. \tag{1}$$

This expression can be transformed to:

$$\overline{\Lambda}_\mathrm{D} = \frac{1}{d}\left(\overline{E} + \frac{\overline{E^2} - \overline{E}^2}{\overline{E}}\right) = L + \frac{\sigma^2(E)/\overline{E}}{d}. \tag{2}$$

* Note that Λ, while being related to Rossi's y, has a much more restricted meaning.

Because 'successive intervals of energy degradation contribute additive amounts to the mean square fluctuation of energy loss' (Fano 1953) the term $\sigma^2(E)/\bar{E}$ is independent of n. Its value is most easily derived in the limiting case of a very small collision number, $\epsilon \ll 1$. The distribution $f(E;\epsilon)$ is then a mere superposition of a delta function in $E=0$, and the collision spectrum (or delta-ray spectrum) $w(E)$

$$f(E;\epsilon)=(1-\epsilon)\ \delta(E)+\epsilon\ w(E). \tag{3}$$

If M_1 and M_2 are the mean and the second moment of the collision spectrum, $w(E)$, one obtains:

$$\bar{E}=\epsilon M_1 \quad \text{and} \quad \overline{E^2}=\epsilon M_2$$

and therefore if one disregards terms of the order of ϵ:

$$\sigma^2(E)/\bar{E}=M_2/M_1. \tag{4}$$

This quantity is the 'energy mean' of the collision spectrum and may be designated by δ_2:

$$\delta_2=\frac{\int E^2\, w(E)\, \mathrm{d}E}{\int E\, w(E)\, \mathrm{d}E}. \tag{5}$$

The index in δ_2 is chosen to distinguish this quantity from the 'number' mean:

$$\delta_1=\frac{\int E\, w(E)\, \mathrm{d}E}{\int w(E)\, \mathrm{d}E}. \tag{6}$$

δ_1 is not used in the present context.

In the usual approximation the collision spectrum $w(E)$ is assumed to be proportional to $1/E^2$, so that:

$$\delta_2=\frac{\int E^2k/E^2\, \mathrm{d}E}{\int Ek/E^2\, \mathrm{d}E}=\left[\frac{E}{\ln E}\right]_{E_{\min}}^{E_{\max}}\sim E_{\max}/\ln\left(\frac{E_{\max}}{I^2}\right) \tag{7}$$

where $E_{\max}$ is the maximum energy transfer in a collision, and $E_{\min}$ is equal to $I^2/E_{\max}$. The contribution of the lower limit of the integration has been neglected in equation (7). Actually the value of δ_2 is somewhat larger due to the contribution of the resonance collisions which are not properly represented by the $1/E^2$-spectrum (Fano 1953):

$$\delta_2=\tfrac{1}{2}E_{\max}/\ln\left(\frac{E_{\max}}{I}\right)+\frac{4}{3}T \tag{8}$$

where $E_{\max}$ is the maximum collision energy transfer and T the mean kinetic energy of electrons in the material.

Whenever $E_{\max}$ exceeds the proper cut-off energy, E_Δ, one has to break off the integration at this value. The formula for δ_2 which can be derived from equation (7) then contains both $E_{\max}$ and E_Δ. Because in this case $E_{\max}$ appears only in the argument of the logarithm, it is a reasonable first approximation to merely substitute E_Δ for $E_{\max}$ in order to account for the cut-off. As far as the relation between the interval length, d, in tissue and the cut-off energy, E_Δ, is concerned, it seems appropriate to assume that E_Δ is the energy of a delta-ray with a mean projected range equal to $d/2$.

Under these conditions the values of δ_2 for intervals from 100 Å to 1 μm range from about 150 eV to 500 eV.

If δ_2 is inserted into equation (2) one obtains:

$$\bar{\Lambda}_D = L + \delta_2/d \tag{9}$$

and with the assumptions mentioned above this is represented by the curves of figure 1.

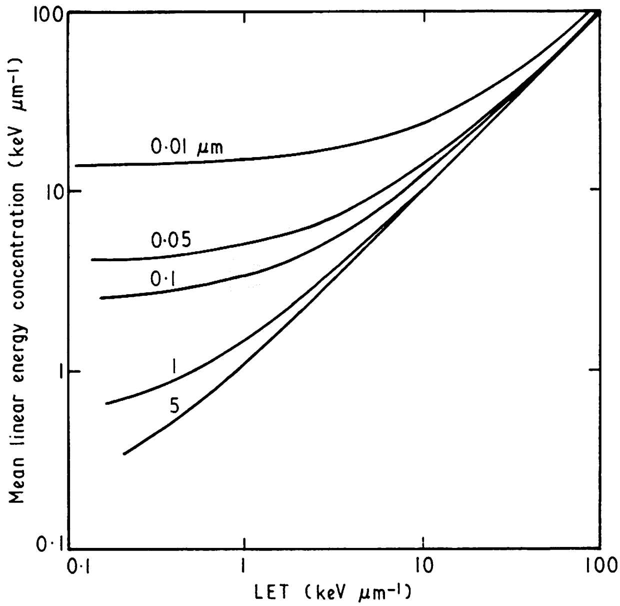

Figure 1. Mean linear energy concentration as a function of ∞. Parameter is the diameter of the region of interest.

The mean value of the linear energy concentration is always larger than LET or its mean value. The excess is due to the clustering of energy deposition along the track; it is most marked for sparsely ionizing radiation and for intervals of less than 1 μm in tissue. In fact in this region the effect of clustering (or straggling) is dominant.

LET is almost irrelevant if it is less than 10 keVμm^{-1} and if one deals with intervals of 100 Å or less. This is borne out in experiments on the inactivation of DNA, and it is generally true for radiation effects on dry materials.

With mammalian cells, on the other hand, there is an appreciable increase of RBE even below 10 keVμm^{-1}. Interaction of energy imparted along the track must therefore extend over fractions of a micrometer. This is in keeping with an analysis of sigmoidal inactivation curves, which shows that neighbouring tracks interact over distances of the order of a micrometer in the inactivation of mammalian cells (Hug and Kellerer 1966).

Figure 1 is also relevant for an understanding of the quality factor, *QF*, as an approximation for the effectiveness of radiation of different LET. The fact that *QF* has been chosen independent of LET below 3·5 keVμm^{-1} is a reflection of the fact that $\bar{\Lambda}_D$, not $\bar{L}_D$, is the parameter which determines the biological effect. A comparison of figures 1 and 2 shows that *QF* is roughly proportional to the mean linear energy concentration over distances of a fraction of a micrometer. This makes sense because the *QF* is related to overall effects on the cellular or intercellular level.

It may be mentioned parenthetically that recent attempts to determine the *QF* with microdosimetry instruments (Baum *et al.* 1969, Bengtsson 1969) should not be looked

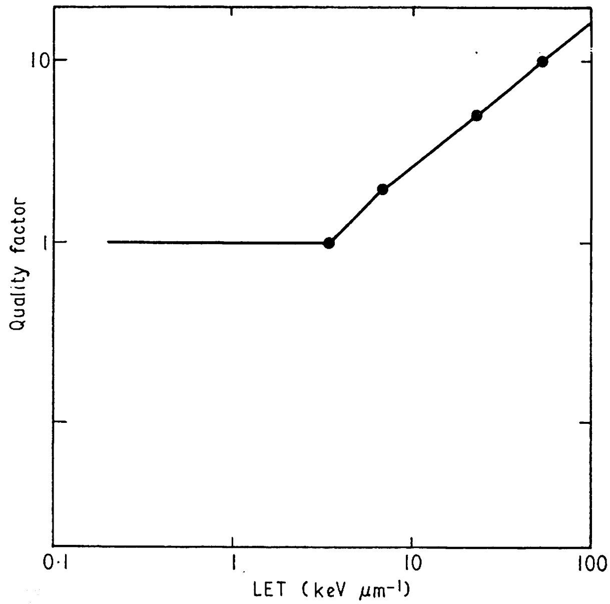

Figure 2. Quality factor as a function of ∞.

upon as approximate methods to derive the LET distribution in order to infer the quality factor. Λ or $\overline{\Lambda}_D$ is the quantity of interest; LET, though formally connected to *QF*, is merely an approximation to the actual energy concentration. Only Λ is experimentally observable and biologically relevant. It is appropriate to determine the local energy concentration over tissue equivalent distances of less than 1 μm in these experiments and simply derive $\overline{\Lambda}_D$ according to equation (1). This mean value can be obtained experimentally if one sums the squares of the pulse sizes in the proportional counter and divides this sum by the sum of the pulse sizes. The effective segment length d is equal to the second moment of the chord length distribution divided by the mean chord length. (This can be shown on the basis of formulae derived on the relative variance of the spectra of energy density (Kellerer 1967, 1969).

While the clustering of energy along the tracks is essential for the biological effect, it does not invalidate the LET concept as such. If one includes the appropriate term δ_2/d the LET values are, indeed, meaningful. LET values refer to a specific cut-off energy, E_Δ, thereby they also refer to corresponding values of δ_2 and d. If the interval size d is taken to be twice the projected range of the electron of energy E_Δ, one roughly obtains 0·01 μm, 0·1 μm, 1 μm, and 5 μm as intervals corresponding to the cut-off energies 100 eV, 1 keV, 4 keV, and 10 keV. Using equations (8) and (9) one finds that for the different cut-off energies one has to replace LET by the following values:

$$\begin{aligned}\overline{\Lambda}_{D,\,100\,\mathrm{eV}} &= L_{100\,\mathrm{eV}} + 15\ \mathrm{keV}\ \mu\mathrm{m}^{-1}\\ \overline{\Lambda}_{D,\,1\,\mathrm{keV}} &= L_{1\,\mathrm{keV}} + 2{\cdot}4\ \mathrm{keV}\ \mu\mathrm{m}^{-1}\\ \overline{\Lambda}_{D,\,4\,\mathrm{keV}} &= L_{4\,\mathrm{keV}} + 0{\cdot}5\ \mathrm{keV}\ \mu\mathrm{m}^{-1}\\ \overline{\Lambda}_{D,\,10\,\mathrm{keV}} &= L_{10\,\mathrm{keV}} + 0{\cdot}23\ \mathrm{keV}\ \mu\mathrm{m}^{-1}.\end{aligned} \tag{10}$$

Note that on the right-hand side of these equations one may as well insert the energy mean, $\bar{L}_{D,\Delta}$, instead of L_Δ. The 'clustering term' depends, at least in our present approximation, only on the cut-off value of the interval size, not on the LET or the nature of the particle.

Instead of introducing the new quantity 'mean linear energy concentration' one might even be tempted to change the definition of L_Δ according to equation (9):

$$L_\Delta = \left.\frac{dE}{dx}\right|_\Delta + \delta_2/d \tag{11}$$

where $(dE/dx)|_\Delta$ is the reduced collision stopping power. That would make L_Δ more than just another symbol for reduced collision stopping power. It would also give some degree of practical usefulness to the concept of energy cut-off in LET theory. The problem of the proper relation between E_Δ and d must, of course, be looked into more closely. The numerical values in equation (10) are only preliminary.

3. Parameters of track structure and the collision spectrum

Considering parameters of track structure, one may tend to think of the average energy, W, expended per ion pair and of δ_1 rather than δ_2. In the preceding section δ_2 has been shown to be the more fundamental quantity. δ_2, not δ_1, determines the mean square fluctuations of energy deposition. A few explanatory remarks on the meaning of the different parameters may help to clarify the relation between W, δ_1, and δ_2.

W together with LET determines the mean number of ionizations produced along a certain interval of the track. In dealing with a condensed medium this is, of course, an ill-defined statement. A sharp distinction between ionizations and excitations is highly questionable in this case. So is the distinction between events of more or less biological effectiveness. W values for a condensed medium are therefore to be used with caution. Moreover W is rather independent of the nature and energy of the ionizing particle, and is therefore not a very significant parameter of track structure.

For a given LET the mean energy transfer in a collision, δ_1 (see equation (6)), determines the mean number of collisions of the primary particle in a certain interval. It also determines the mean spacing, $\bar{l} = \delta_1/L$, between successive collisions. It is, however, important to note that δ_1 depends critically on the initial part of the collision spectrum. This initial part is not well-known at present. The experiments of Rauth and Simpson (1964) indicate that δ_1 has a value of approximately 60 eV for 20 keV electrons. Whether the same value can be assumed for electrons of higher energy or for heavy charged particles is an open question and has to be decided by further experiments on the collision spectrum. In radiobiology appplications δ_1 is not the physical parameter of chief interest. The plasma oscillations induced by the ionizing particle have supposedly little biological effect, while the value of δ_1 is, however, strongly influenced by these events of small energy transfer. Let us specifically assume that only 'ionizations' are considered as events. A conservative estimate is that half of all primary collisions do not lead to ionizations. The value of δ_1 measured in gas on the basis of ionizations alone must therefore be at least twice as large as the true value δ_1 in the condensed medium. This is in agreement with values of about 120 eV given by Hutchinson and Pollard (1961), while measurements by Sommermeyer and Dresel (1955, 1956) in the gas phase indicate lower values of δ_1.

Apart from being somewhat poorly defined δ_1 may also depend appreciably on the velocity of the ionizing particle. Calculated collision-spectra (Bichsel 1969, Choi and Merzbacher 1969) clearly indicate this dependence. The applicability of δ_1 is further limited by the fact that one can distinguish primary collisions only when their mean spacing, δ_1/L, is large as compared to the mean range of delta-rays or the mean cluster size. This requires that L is well below 10 keV μm^{-1}.

The bearing of δ_2 on track structure has been discussed in detail in the preceding section. The underlying fact may be illustrated by a simple statement. For larger intervals the distribution of energy deposition by a heavy charged particle is Gaussian. A Gaussian distribution of the same width results if the particle transfers energy in statistically independent events all of size δ_2. The value of δ_2 is rather insensitive to the initial part of the collision spectrum. It also makes little difference whether one deals with all collisions or with ionizations alone. From Fano's considerations on the fluctuation in the number of ions (Fano 1947) one derives that δ_2 increases only by $W/2$ (≈ 17 eV) if one determines its value on the basis of ionizations alone. Thus δ_2 in contrast to δ_1 can be determined very well from experiments in the gas phase. Since δ_2 depends strongly on the cut-off at high energies (see equation (8)) one must use devices which measure the energy imparted to the volume of interest, not the energy lost by the ionizing particle. Straggling experiments with thin foils usually yield values of $\delta_2 = \sigma^2(E)/\bar{E}$ which do not reflect a cut-off since one measures the energy loss of the primary particle, not the energy locally imparted. Straggling results from semiconductors (Maccabee 1968) are at present of limited significance to radiation biology since the equivalent layers are still too thick. Moreover the geometry of a foil cannot readily be compared to that of a cylindrical or spherical volume. For this reason the experiments with wall-less proportional counters by Rossi and Gross and by Glass, Roesch and Braby, which are being reported at this conference, are most useful for a determination of the effective δ_2 in microscopic volumes of different sizes. W. Glass *et al.* have computed numerical values of $\delta_2 = \sigma^2(E)/\bar{E}$ from experiments with protons of a few MeV; these values are consistent with the values derived in the preceding section.

The track of an ionizing particle is, of course, much too complicated to be described by a few parameters. The next step is therefore the question of the complete collision spectrum, $w(E)$. One answer to this question is given by observations on the electrons ejected from very thin foils. The answer, however, is incomplete, since one observes the kinetic energy of the electrons, not the total energy transfer including the binding energy. Thus it is not an indication of the applicability of the $1/E^2$ model, if one finds that, with the exception of the Auger peaks, the energy of the ejected electrons is distributed as $1/E^2$. On the contrary, this experimental finding supports the assumption that the collision spectrum is steeper than $1/E^2$ at low energies. Another approach is the observation of relative frequencies of ion clusters of different size in cloud chambers. The possibilities and the limitations of these observations are discussed in this volume in detail.

A third and very significant method is the use of straggling experiments in thin foils or in small gas gaps or volumes. The application of this method to the analysis of collision spectra has been limited in the past, because the corresponding mathematical problem, the compound Poisson process (Kellerer 1968a), had only been solved in the limit of many collisions and under the assumption of the $1/E^2$-spectrum or its relativistic modification. In the remaining cases rough numerical estimates or Monte Carlo methods had to be applied.

These restrictions have contributed to the fact that straggling distributions (i.e. distributions of the energy loss) have normally been determined for high collision numbers only, where the classical theory with its corrections according to Blunck and Leisegang (1950) and to Shulek *et al.* (1967) is adequate. Very little information on the collision spectrum can be derived from these many-collision experiments. On the other extreme one has tried to work with foils so thin that multiple collisions could be disregarded. This method is severely limited by the difficulty of controlling the purity and thickness of very thin foils and by the infrequency of near collisions and the resultant poor statistics in the high energy part of the collision spectrum. In the experiments of Rauth and Simpson (1964), for example, no significant information is available in the important region between 100 eV and 1 keV.

Straggling experiments with intermediate collision numbers, however, can now be analysed. Recently a computer programme has been developed which derives straggling distributions for arbitrary collision spectra and arbitrary collision numbers. The only condition is that the collision spectrum $w(E)$ is constant throughout the interval of interest. The programme (a new realization of an attempt which has been made here in Cambridge 40 years ago by E. J. Williams 1929) is based on the fact that energy loss in successive intervals is statistically independent. Starting from the approximation for extremely thin layers (see equation (3)) straggling distributions are derived in a series of successive convolutions; each of the successive distributions belongs to doubled thickness and doubled mean energy loss. The convolutions are executed on a logarithmic scale, since in certain cases the collisions spectrum covers so many orders of magnitude that use of a linear scale is not only inefficient but out of the question. This programme is available in Algol 60 and in Fortran IV. It has been published together with a theoretical analysis of the problem and its implications to radiobiology (Kellerer 1968a). An approach on the same line has independently been chosen by W. Roesch (1968).

The computer programme derives straggling distributions from collision spectra; it does not derive collision spectra from straggling distributions. One can obtain useful information on the collision spectrum by comparative curve fitting (Kellerer 1967). But ideally one would like to reverse the process. This is indeed possible. In mathematical terminology: a solution of a compound Poisson process uniquely determines the characteristic spectrum of the compound Poisson process. The reason can for example be understood from Fano's remarks on the Laplace-transform and the additivity of straggling (Fano 1953). These remarks hold for the characteristic functions (Fourier transforms) of a distribution as well as for the Laplace transform. It is therefore not necessary to repeat the argument here, in much detail.

If $\phi(\tau)$ is the characteristic function of the straggling distribution $f(E; n)$, then the characteristic function to the distribution $f(E; k.n)$ which belongs to a mean energy loss k-times as large is equal to $\phi(\tau)^k$; convolution reduces to mere multiplication for the characteristic functions. The relation holds for arbitrary non-negative values of k. Therefore it can also be applied to reduce the mean value. The only condition is that one must choose a proper continuous representation of $\phi(\tau)$ in the complex plane instead of deriving it modulo $\exp(2\pi i)$. Otherwise the function $\phi(\tau)^k$ would not be uniquely defined. This idea has been used to develop a computer programme for 'devolution' as well as convolution of given distributions. The programme (available in Fortran IV) uses the fast Fourier transform algorithm of Cooley and Tuckey (1965). Given an experimental straggling distribution to a certain mean energy loss (layer thickness) the straggling distribution to a larger or smaller mean energy loss (layer thickness) is computed. In choosing this mean value sufficiently small one directly obtains the collision spectrum.

With increasing mean values the solutions of the compound Poisson process converge to normal distributions regardless of the characteristic spectrum. In view of this fact it may seem unlikely that the underlying spectrum can be recovered from an arbitrary solution. Indeed the method is limited by the fact that small errors in the input data lead to considerable blurring of the resulting curve as the computer programme goes back to smaller mean values. Thus the method is applicable only if one either starts with moderate mean collision numbers or with extremely well-defined distributions, which are indeed 'infinitely divisible' (Gnedenko and Kolmogorov 1949, Linnik 1964). This is not a limitation of the mathematical procedure. It is, in fact, one of the main advantages of the programme that the results clearly indicate the range of different functions equally compatible with a particular result. Though it is beyond the scope of this paper to go into details of technique the procedure has been mentioned because it is felt that its existence may stimulate further experimental work on the analysis of the collision spectrum of charged particles.

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