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CONTENTS

MICRODOSIMETRY AND THE THEORY OF STRAGGLING

A. M. KELLERER
STRAHLENBIOLOGISCHES INSTITUT DER UNIVERSITÄT, MUNICH
AND
INSTITUT FÜR BIOLOGIE
DER GESELLSCHAFT FÜR STRAHLENFORSCHUNG
NEUHERBERG
FEDERAL REPUBLIC OF GERMANY

Abstract

MICRODOSIMETRY AND THE THEORY OF STRAGGLING. Theoretical developments in microdosimetry (the determination of the spatial distribution of energy deposition on a microscopic scale) are described. It is pointed out that microdosimetric theory is equivalent to LET theory plus straggling theory. The influence of each of the different random factors (varying LET, varying track length, varying number of primary collisions and varying energy transferred in primary events) can be expressed as a relative variance and these relative variances can be summed to give the relative "variance" of the single-event spectrum. The track-length variation and the variation in the number of primary events will often not be important. Although the LET variation may be quite important, it appears that the energy spectrum of primary collisions will usually make the decisive contribution.

This paper describes studies following those concerned with the biological application of microdosimetry, with the mathematical scheme of functions needed for this purpose, and with a first theoretical determination of some of these functions [1]. The previous theoretical calculations, based on Monte Carlo methods, were in good agreement with the experimental data. However, new improvements in the experimental technique made it desirable to develop accurate theoretical methods. While more refined and, necessarily, more abstract computations are applied, one should also attempt a clearer understanding of the different factors involved. This paper therefore, outlines a general theoretical investigation into the spectra of local energy density. The basis is Rossi's concept of local energy distribution [2]; the work reported was done partly in his laboratories at Columbia University.

This work is divided into two sections: First, the different random factors which make up the distribution of local energy density are briefly discussed. A formula is then given which comprises all these factors. This formula is the basis for the numerical calculations and has been used successfully in the past. It is, however, rather complicated and does not give a simple idea as to the relative contributions of the different factors to the shape of the local energy spectra. Because of this difficulty, a relation of remarkable simplicity is presented which connects the variance of the microdosimetry spectra with the variance of the individual random factors. This relation leads to the second part and to a point which is central to this paper. It turns out that, except for large volumes

and very densely ionizing radiations, one factor dominates. This is the wide range of energy loss in primary collisions. Its influence on the shape of local energy density spectra is in many cases decisive. But even in cases where it is not the dominant factor, it is still the one which presents the most serious difficulties – difficulties as far as physical knowledge and mathematical evaluation are concerned. At the same time, this is the factor which is most characteristic for the new aspects brought up by microdosimetry. Fluctuations of energy deposition are, however, by no means a new subject. It may be said that microdosimetry is LET theory plus straggling theory.

While the connection between LET studies and microdosimetry is one of the general topics discussed by this Panel, the alternative aspects – microdosimetry and straggling – are stressed here.

(a) Interplay of different random factors

To elucidate the different random factors responsible for the local energy density spectra, below is briefly described the chain of events which takes place in the measurements of local energy density spectra:

A tissue-equivalent proportional counter is exposed to a dose D of ionizing radiation. The number of passages of ionizing particles through the counter is a random variable. Discussion is confined to the simple case that one has to deal only with complete passages of the ionizing particles through the sensitive volume. The ionizing particle is assumed to lose only a small fraction of its kinetic energy in a passage. This loss of energy occurs in statistically independent "primary collisions". The expected number of primary collisions is proportional to the track length and to the stopping power of the particle. Therefore, one has to know the track length distribution as well as the LET distribution. For a given track length and a given value of LET the number of primary collisions is still a random variable. Not only is the number of primary events a random variable, but also the size of these events varies, i.e. the amount of energy transferred in a primary collision is distributed statistically. Actually, the resulting δ-ray may have a range long enough to leave the sensitive volume; for the time being, however, this is not considered, but it is assumed that all energy is deposited locally. In other words, all primary events are considered to be point events. Corrections for the above simplification can be applied afterwards.

A certain amount of deposited energy will lead to a number of ionizations which, in turn, is subject to statistical fluctuations (these may be called "Fano fluctuations", since Fano gave the first theoretical analysis of the problem [3]). The next step is that each of the electrons freed in an ionization is accelerated towards the anode. At the end of its path it generates a varying number of secondary ionizations which in turn multiply and thus contribute to the pulse which corresponds to the passage of the ionizing particle. Actually there are also some less essential factors like amplifier noise or imperfect function of the counter. These factors which, in principle, can be reduced by experimental technique, need not be dealt with here.

Actually, the discussion may also be restricted to the case of the single-event spectra $f_\Delta(Z)$, since from these spectra the distributions $f(Z;D)$[1] for different doses D can be calculated (see formula (6)). To arrive at a complete formula for the distribution of local energy density the following questions have to be asked:

1. What is the distribution $s(\ell)$ of track length ℓ in the critical volume? In the special case of a sphere: $s(\ell) = 2\ell/d^2$ (where d = diameter).
2. What is the distribution r(L) of LET? The distribution relative to the track length is requested.

Factors 1 and 2 combine to make up the probability distribution t(E) of the expected value E to be deposited in the passage. E is the energy which would be laid down if there were no straggling.

3. What is the distribution $p(\nu)$ of the number ν of primary collisions along the track? This number is distributed according to Poisson, since the primary collisions are statistically independent. If δ_1 is the mean energy transferred in a primary collision,

$$p(\nu) = e^{-E/\delta_1} \cdot (E/\delta_1)^\nu/\nu! \qquad (1)$$

4. What is the distribution w(e) of energy e transferred in a primary collision? This function is proportional to $1/e^2$ for energies large compared to the binding energy of the electrons. The shape at lower energies is, however, quite important for the microdosimetry spectra.

It will be seen that the determination of the distribution w(e) and its "number" and "energy" mean δ_1 and δ_2 is the central problem in the theoretical deduction of the spectra of local energy density. It is also decisive for all considerations on "straggling", and it is here where microdosimetry and the conventional straggling theories of Bohr [4], Landau [5], Symon [6] and Vavilov [7] meet. These are the probability distributions which determine the theoretical spectra. To continue, what are the factors which are introduced by the experimental method?

5. How many ionizations are produced by the energy e? The expected number of ionizations is e/W (W = 32 eV). Actually there are fluctuations. The simplest assumption would be that λ is distributed according to Poisson. Theoretical considerations (Fano [3]) indicate that the distribution $\varphi(\lambda; e)$ is not Poissonian but of a variance approximately half of the Poissonian. Not much is known beyond this. This random factor will turn out to be of relatively minor importance.

6. What is the distribution $\gamma(\zeta, \lambda)$ of pulse height ζ brought about by λ initial electrons? If (as preliminary experiments indicate for spherical counters) $\gamma(\zeta)$ is exponential, the distribution of pulse height brought about by λ initial electrons is:

$$\gamma(\zeta_i \lambda) = e^{-\zeta} \zeta^{\lambda-1}/(\lambda-1)! \qquad (2)$$

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[1] This is a preliminary notation as long as no definitive notation has been established in microdosimetry. $f_\Delta(Z) \cdot dZ$ is the probability that an increment of local energy density between Z and Z + dZ is produced by an absorption event. $f(Z; D) \cdot dZ$ is the probability that a local energy density between Z and Z + dZ results if the dose D is applied. The functions depend on the size of the critical volume and on the radiation quality; they correspond to Rossi's $P(\Delta Z)$ and $P(Z)$.

This factor is also rather unimportant. Moreover, a computer programme has been developed for correction of the experimental spectra.

With these distributions the formula can, in principle, be constructed for the local energy density spectra. There is a need, however, not only for the elementary distributions, but also for higher order distributions. While $s(\ell)$, $w(e)$, etc. are distributions for one event, it is also necessary to know the distributions which result if several events occur. One obtains these distributions by "folding" the elementary ones. This operation, which is also called 'convolution', poses the central mathematical problem in straggling theory as well as in the computation of microdosimetry spectra. In the past it has led to considerable difficulties and has therefore attracted much interest. We have developed a computer programme which solves this problem (see (b) of this paper). Thus, for the moment, it may be assumed that the resulting distributions are known. In the formulae, however, a mathematical symbol is needed for this operation and its repeated application. For this purpose, a star is used. If $w(E)$ is the probability distribution of energy spent in a single collision, the distribution of energy spent in two collisions is called $w^{*2}(E)$. As may be seen:

$$w^{*2}(e) = w(e)^* w(e) = \int_0^e w(e-x)\cdot w(x)dx. \tag{3}$$

More generally the n-th 'folding power'[2] of $w(E)$ is designated by $w^{*n}(e)$. This is the distribution of energy spent in exactly n collisions:

$$w^{*n}(e) = \int_0^e w^{*(n-1)}(e-x)\cdot w(x)\,dx,$$

or

$$= \int_0^e w^{*(n-\nu)}(e-x)\cdot w^{*\nu}(x)dx \tag{4}$$

or, in general form,

$$= w^{*(n-\nu)}(e)^* w^{\nu}(e)$$

If this convention is applied also to the other distributions, one may write the complete formula for the single event spectrum $f_\Delta(Z)$:

$$f_\Delta(Z) = \sum_{\nu\lambda} \int_{E,e} t(E)\cdot e^{-E/\delta_1} (E/\delta_1)^\nu/\nu!\cdot w^{*\nu}(e)\cdot\varphi(\lambda_1 e)\cdot\gamma^{*\lambda}(Z)dE\cdot de \tag{5}$$

[2] This analogy to multiplication is based on the fact that convolution reduces to mere multiplication if one goes over to the characteristic functions of the distributions.

Each term corresponds to one of the random factors. It may be mentioned again that the last two terms (indicated by Greek letters) are brought in by the experimental method, namely by the use of proportional counters. For theoretical determination of the spectra of local energy density these terms are to be dropped.

At this point the formula which allows deduction of the dose-dependent spectra from the single event spectrum may be added:

$$f(Z;D) = \sum_{\nu=0}^{\infty} e^{-D/\Delta_1} \frac{(D/\Delta_1)^{\nu}}{\nu!} \cdot f_{\Delta}^{*\nu}(Z) \qquad (6)$$

where Δ_1 = mean local energy density produced in one absorption event.

The formula is easily understood. The first, Poissonian, term gives the probability for exactly ν absorption events while $f_{\Delta}^{*\nu}(Z)$ is the distribution of Z under the condition that ν absorption events take place.

A detailed discussion of these formulae is not necessary here (for reference see [1]). They may just serve to show what kind of problems one encounters. The mathematical problem is that of finding the distributions which are generated by repeated folding from an initial distribution. The solution of this problem is discussed in the next section.

An additional problem is that of determination of the LET distribution and of the distribution w(e) of energy transfer in primary collisions. The LET spectra are well discussed, and in many special cases they can be given numerically. The distribution w(e), however, presents serious difficulties. This is discussed at the end of the paper.

First, a relation is mentioned which enables comparison of the influence of the different random factors. Specifically it brings out the importance of the "straggling" term.

Each of the random factors: varying LET, varying track length, varying number of primary collisions, and varying energy transferred in primary events, has the effect of broadening the spectra of local energy density. The variance of the local energy density spectra is a convenient measure for the combined influence of the different factors. A suitable dimensionless term is $V = \sigma^2/\overline{Z}^2$; we call it relative variance. V is small if the fluctuations are small as compared to the mean. It is well known that for distributions with $V \ll 1$ (specifically for Gaussian distributions) the variance due to different perturbations ("weak" random factors) simply adds. A similar fact is not to be expected for the spectra $f_{\Delta}(Z)$ of local energy density, where $V \gg 1$, i.e. where the fluctuations are even bigger than the mean values. Surprisingly it turns out that, nevertheless, a simple additive relation holds. One may take each individual random factor, neglect all the others, and calculate the resulting relative variance. If this is done for all different factors, all the resulting terms may be simply summed up; thus one obtains the actual value for the relative variance of $f_{\Delta}(Z)$. There is just one cross term, namely for LET and track length.

There are certain limitations, since this relation strictly holds only under simplifying assumptions, e.g. complete passages and treatment of the primary collisions as point events. Still it is an excellent tool in practical cases and gives the right order of magnitude for the contribution of the different factors.

The main points can be explained by writing down the relation in its most simple form without proof. It may be mentioned that it can easily be extended to the dose-dependent spectra. In passing, it may also be remarked that the whole concept is closely related to similar notions in the analysis of dose-effect curves. There, in addition to physical factors, biological parameters contribute to the variance of the curves, but knowledge of the relative variance of the local energy density spectra is sufficient to deduce a lower limit for the size of the sensitive area [1].

It can be shown that, for the relative variance V of the single-event spectrum $f_\Delta(Z)$, the following relation holds:

$$V = V_{Track} + V_{LET} + V_{Track} \cdot V_{LET} + V_{Straggling} + V_\varphi + V_\gamma \qquad (7)$$

$$= 1/8 + 9/8\, V_{LET} + \frac{\delta_2}{\Delta_1} + \frac{\sim 16[eV]}{\Delta_1} + \frac{\sim 32[eV]}{\Delta_1}$$

where Δ_1 is the mean energy laid down in an absorption event (i.e. passage of an ionizing particle through the critical volume), δ_2 is the "energy" average of energy transferred in primary collision. For V_{track} the value 1/8 is inserted which corresponds to a spherical volume[3]. δ_2 depends on the type of radiation, but as a general rule it may be stated that it is equal to several hundred eV. In the special case of 5.75 MeV α-particles (see Rossi and Rosenzweig [16], and also (b) of this paper) $\delta_2 = 430$ eV.

From this equation many observations can be deduced. To mention a few: First, the last two terms, which are due to the Fano fluctuations and to the statistics of electron avalanche formation, are always small compared to the straggling term. Their combined contribution is approximately 45 eV/Δ_1, that is, 10% of the straggling contribution to the variance (5% in terms of the curve width; thus these two factors are not a very serious limitation of experiments in the gas phase. For solid state detectors they are still less important, because there the mean energy needed for an ionization is much smaller than 32 eV).

Second, it may be observed that the influence of the track-length variation is not decisive as long as Δ_1 is less than several keV. Thus, for sparsely-ionizing radiation the shape of the counters is of minor importance. One may expect to find similar results with spherical and non-spherical counters.

Naturally, the LET term may be quite important. It will be of great interest to calculate V values for LET distributions. Actually this is a familiar problem. It is usual to compute two different averages of a

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[3] Usually V_{Track} is somewhat larger than 1/8 if the volume is not spherical. Perhaps it is possible to prove this fact in general for non-spherical convex volumes.

LET distribution, namely $\bar{L}_D$, the dose average (equal to m_2/m_1, where m_2 and m_1 are the second and the first moment of the distribution), and $\bar{L}_T$, the track average (equal to m_1). These two averages, however, determine the relative variance of the LET distribution[4]:

$$V_{LET} = \frac{\sigma^2}{m_1^2} = \frac{m_2 - m_1^2}{m_1^2} = \frac{\bar{L}_D}{\bar{L}_T} - 1 \qquad (8)$$

A corresponding relation can be given for the single-event spectra $f_\Delta(Z)$ of local energy density:

$$V = \frac{\bar{Y}_D}{\bar{Y}_P} - 1 \qquad (9)$$

where $\bar{Y}_D$ and $\bar{Y}_P$ are the dose and the number average of the event size.

Comparing the values V and V_{LET} one can judge the relevance of the LET distribution for the shape of a local energy distribution. The difference between V and V_{LET} is due to the other factors (like straggling or track length distribution). If V and V_{LET} are not very different, the local energy density spectrum is mainly determined by the LET spectrum. This can be the case especially for neutron irradiation and volumes which are small enough that most absorption events are due to complete passages. In this case the mean energy Δ_1 transferred in an absorption event equals several keV and the straggling term $V_{Straggling} = \delta_2/\Delta_1$ is very small. Therefore, it is quite justified that Cashwell, in his theoretical determinations of the event spectra for neutron radiation, has completely neglected the straggling effects.

Take a practical example: For 0.5 μm diam. and 5.7 MeV neutrons the experimental data are $\bar{Y}_D$ = 61.8 keV/μm and $\bar{Y}_P$ = 12.5 keV/μm (Biavati, Rossi, Boer [8]); this leads to V = 3.95. Bewley (see his paper in this report) gives the values $\bar{L}_D$ = 87 keV/μm and $\bar{L}_T$ = 20 keV/μm for 6 MeV neutrons. The relative variance of the LET distribution is therefore V = 3.35. Obviously, the LET distribution is the dominant factor for the shape of the local energy density spectra. While the fine structure of the LET distribution is, of course, washed out in the microdosimetry spectra, the main shape is the same in both cases. The difference between V and 9/8 V_{LET} (see formula (7)) is only 0.3. Of this difference 0.125 is due to the track-length distribution in the spherical counter, and the rest represents the straggling and the influence of the incomplete passages.

For ^{60}Co γ-rays and a tissue-equivalent diameter of 1 μm the experimental values $\bar{Y}_D$ = 1.24 keV/μm and $\bar{Y}_P$ = 0.262 keV/μm are given [10]; the corresponding relative variance is V = 3.73. The spectrum which has been computed theoretically [1] has a relative variance V = 3.5. The

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[4] This is the reason why both 'averages' have to be used in LET theory. $\bar{L}_T$ is the true mean, while in addition $\bar{L}_D$ determines the variance of the LET distribution, $\sigma^2 = (\bar{L}_D - \bar{L}_T)\cdot\bar{L}_T$.

difference in the two values reflects a somewhat bigger tail of the experimental curve at high local energy densities. This may either be due to wall effects in the proportional counter or to the fact that in the computations δ-rays with energy beyond 3.5 keV have been excluded. Most of the variance of the ^{60}Co γ-spectra goes back to the straggling term. The contribution of the track length spectrum is quite unimportant. It is remarkable that, with its value of 0.125 (see formula (7)), it contributes even less than Fano fluctuations and counter resolution (45 eV/262 eV = 0.17). This means that, for this type of radiation, and for small volumes, it is not essential that a well-defined spherical counter is used. This will simplify future experiments, specifically those with "wall-less" chambers. The LET distribution cannot be neglected, but for a cut-off of 3.5 keV the relative variance of the LET spectrum is smaller than one. Cormack [11] gives $\overline{L}_D$ and $\overline{L}_T$ values for ^{60}Co γ-radiation for a cut-off of only 0.1 keV. In this case the relative variance of the LET spectrum is very large. But as Cormack (see his paper in this report) remarks, much higher cut-offs are relevant for the microdosimetry spectra. With a higher cut-off the LET distributions become more narrow.

(b) Some further remarks concerning straggling

The spectrum w(E) of energy transferred in primary collisions presents the most formidable difficulty in theoretical microdosimetry. It is, therefore, justified to discuss this distribution separately. As soon as w(E) and its folding powers $w^{*\nu}(E)$ are known, the other factors can be included, and the complete event spectra may be calculated. Thus, discussion may be reduced to the most simple case. An ionizing particle traverses a thin layer of matter, and we ask for the distribution of energy loss. In abstract form this distribution can be written down quite easily:

$$f(E) = \sum_{\nu=0}^{\infty} e^{-\overline{E}/\delta_1} \, (\overline{E}/\delta_1)^{\nu}/\nu! \cdot w^{*\nu}(E) \qquad (10)$$

where $\overline{E}$ is the mean energy loss and δ_1 is the mean of w(E). The actual solution is, however, a highly complicated problem and it is the central point of the theory of energy loss fluctuations. Before the different ways to a solution are discussed, a few simple remarks are made below which may illustrate the problem.

If an ionizing particle traverses a thin foil it may lose a small fraction of its kinetic energy. This loss occurs in statistically independent primary collisions according to Eq. (10). Assume that in all primary collisions the energy δ_1 is laid down. Then obviously the relative variance of the curve depends on the mean number of collisions. If the mean number of collisions is large, the element of chance is reduced and the curve becomes relatively narrow. In fact, one can show that, in this case, the relative variance $V = \delta_1/\overline{E}$, i.e. V is inversely proportional

to the mean number $\bar{E}/\delta_1$ of collisions. From Eq.(7) it can be seen that actually $V = \delta_2/\bar{E}$. Thus V is indeed inversely proportional to the mean number of collisions but its value is increased by a factor δ_2/δ_1 (which is typically between 5 and 10). δ_1 and δ_2 are the two different "mean" values of w(E).

$$\delta_1 = \int E\, w(E)dE \quad \text{and} \quad \delta_2 = \frac{\int E^2 \cdot w(E)dE}{\int E \cdot w(E)dE} \tag{11}$$

Thus, the variance of the straggling curves is mainly determined by the shape of w(E) and very little by the Poissonian fluctuations of the number of primary events. Therefore any description by a pure Poisson process (i.e. by equal primary events which are distributed according to Poisson) is extremely unrealistic. If a comparison is to be made at all, one must choose an event of size δ_2 (several hundred eV) and neither δ_1 (approximately 60 eV), which is the true mean collision energy, nor W (32 eV), the mean energy per ionization. The latter assumptions would lead to curves which are much too narrow. The relation:

$$V = \delta_2/\bar{E} \quad (\text{or } \sigma^2 = \delta_2 \cdot \bar{E}) \tag{12}$$

is practically quite important. If δ_2 is known, one can immediately deduce the variance of a straggling curve (and also the width, if the curve is not very asymmetrical). Thus, experimental curves can easily be checked. Conversely, experimental determination of the variance of the energy loss leads directly to the value δ_2 and thus to some information on the distribution w(E).

In experiments on energy loss fluctuations the results are usually compared with the distributions derived by Vavilov [7]. In the non-relativistic case these distributions are based on the assumption that $w(E) \sim 1/E^2$. The maximum energy E_{max} of δ-rays is equal to $4m/M \cdot E$ (where E is the kinetic energy of the ionizing particle and m/M the mass ratio). To arrive at the right value of the stopping power the spectrum is extended down to a minimum energy $E_{min} = I^2/E_{max}$. With these assumptions one can deduce the theoretical value of δ_2. This is important, because, in general, it should be the first step to check the applicability of the Vavilov distributions by comparison of the experimental and theoretical variance. From formula (11) it follows that:

$$\delta_2 = \frac{\int dE}{\int \frac{1}{E} \cdot dE} = E_{max}/(2 \ln(E_{max}/I)) \tag{13}$$

(with $E_{max} >> E_{min}$).

Several factors are responsible for possible differences between experimental values $\delta_2 = V \cdot \bar{E}$ and the theoretical δ_2. As can be seen from formula (7) the Fano fluctuations and the resolution of the pro-

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portional counter lead to an increase of about 45 eV for measurements in the gas phase. While this is an error brought in by the experimental method, the resonance collisions are responsible for a true increase in δ_2. The magnitude of this increase depends on the atomic composition of the irradiated medium and on the velocity and mass of the incident particle. There is, however, also a factor which decreases the experimental value of the variance of the energy loss distribution. This is the loss of δ-rays. If the maximum range of δ-rays is comparable with the width of the critical gap or the thickness of the sensitive layer of a solid state detector this factor cannot be neglected. The fact that the deviations from the theoretical model may balance out to a certain extent makes it hard to judge the validity of the $1/E^2$ relation in practical cases. More refined analysis has to be used, then, and a few remarks are added in this direction.

Until now exact computation of the energy loss distributions has been possible in the special case $w(E) \sim 1/E^2$ only (or in its relativistic modification). The classical formula is valid for a free electron gas, but it is quite unrealistic for energies in the order of magnitude of the electron binding energies. Thus the classical theory is restricted to cases where E_{max} (the maximum δ-ray energy) is very large compared to the binding energies. Under these conditions the classical theory is well verified by experimental results [12]. In all other cases corrections have to be applied. These corrections take into account the increase in relative variance of the curves brought about by the distant collisions; but one has to use rather crude approximations, and cannot take into account the exact shape of the spectrum w(E) at small energies (for reference see [12, 13]). Therefore the theoretical distributions are not applicable to cases where the mean number of primary collisions is small; these, however, are the cases of interest in radiobiological applications.

Even within their limits the conventional mathematical methods are not completely satisfying. While Bohr and Landau have deduced the solutions for special ranges of $\overline{E}$ only, Vavilov has found a general analytical solution; this solution, however, is complicated enough that it in turn has to be evaluated by a special computer programme (Seltzer and Berger [13]). Direct numerical evaluation seems to be more useful in some respects. If it is desired to work with realistic spectra w(E), this method is the only way. In microdosimetry applications one is forced to cut off the w(E) spectra at higher energies. This corresponds to the loss of δ-rays which have a long enough range to leave the critical volume. Because of this cut-off the shell corrections in w(E) are of still greater importance. One practically has to deal with the distant collisions alone. The corrections as well as the cut-off for energetic δ-rays are, of course, equally important in straggling experiments with thin solid state detectors or narrow gas gaps. For these reasons a computer programme has been developed for the solution of Eq. (10). This programme works for arbitrary distributions w(E) and has no limitations as to the range of $\overline{E}$ (or, in the usual terminology, the range of κ). The programme can also be used to compute the dose dependent local energy distributions $f(Z; D)$ from the single-event spectra $f_1(Z)$. This is due to

the fact that Eqs (6) and (10) are of the same structure; they both describe a composite Poisson process.

This is not the first attempt towards a purely numerical solution of Eq. (10). Williams treated this problem in 1929; naturally, his results are rather crude. Monte Carlo methods have been applied repeatedly. One and the same Monte Carlo Programme has been used [1] to calculate energy loss distributions, single-event spectra $f_\Delta(Z)$ and dose-dependent spectra $f(Z;D)$ of local energy density. Biavati [14] has worked out a programme which attacks Eq. (6) in a straightforward way to compute the distributions $f(Z;D)$ from $f_\Delta(Z)$. While this programme is much more exact than the Monte Carlo methods, it involves an extremely large number of computations and is therefore restricted to a limited range of D. While this limitation is usually not too serious for Eq. (6), it turns out to be prohibitive in the case of the straggling problem. New methods had to be found to avoid the relative inaccuracy of the Monte Carlo methods and still obtain a wide range of solutions for any primary distribution.

For an understanding of one of the main points of the new programme the following observation is necessary: on a linear scale of E the straggling curves become successively broader with increasing mean energy E. On a logarithmic scale the curves become more narrow with increasing $\overline{E}$. Both scales, therefore, present difficulties if they are used in the numerical treatment. It turns out, however, that with a square root scale of E the 'width' of the curves is independent of $\overline{E}$. With increasing $\overline{E}$ the curves even approximate constant shape. This follows directly from relation (12). Thus the square root scale of E is the one to be chosen for numerical calculations. It is also very illustrative for presentation of the results. In Fig. 1 the different ways of plotting are compared. These are calculated energy loss distributions for 5.75 MeV α-particles in water.

As a second central point of the programme it may be mentioned that it is not at all necessary to compute all the successive convolutions $w^{*\nu}(E)$ of the initial distribution. Instead, the energy loss distribution $f(E)$ is first computed for a mean energy loss which corresponds to one or less primary collisions in the mean. For this purpose Poisson superposition of the first eight "folding powers" of $w(E)$ is quite sufficient. For example:

$$f_1(E) = \sum_{\nu=0}^{8} e^{-1}/\nu! \cdot w^{*\nu}(E) \qquad (14)$$

is the energy loss distribution which corresponds to one primary collision in the mean. To obtain the energy loss curves for higher mean energies one can start directly from $f_1(E)$, and need not go back to $w(E)$ at all. By a first convolution $f_1(E)*f_1(E)$ one obtains the curve for $E = 2\cdot\delta_1$, by a second convolution the curve for $4\cdot\delta_1$ and so on. By only 10 successive convolutions an energy is arrived at which corresponds to 1024 primary collisions. The computation times are therefore short. Of course, one

is not limited to energies which correspond to integral numbers of primary collisions. Poisson factors in Eq.(14) may be chosen with an arbitrary mean.

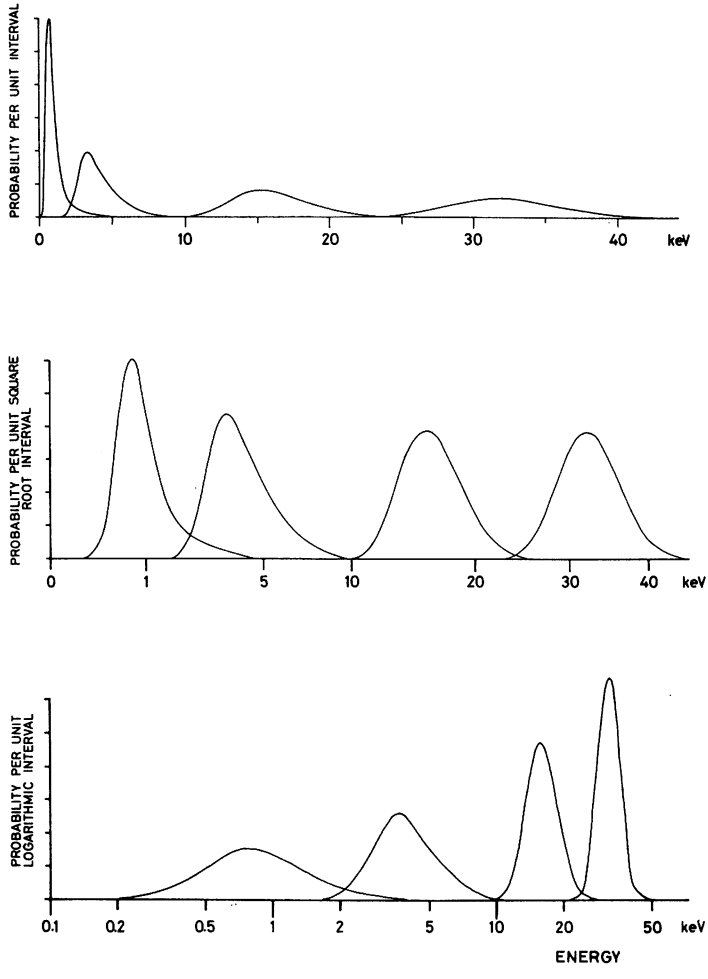

FIG.1. Energy loss distributions for 5.75 MeV α-particles in water. The mean energies are 1 keV, 4 keV, 16 keV, and 32 keV. The corresponding distances in water are 120Å, 500 Å, 0.19 μm and 0.38 μm

The technical details of the computations are not gone into further here. Instead, Fig. 2 gives a more exact representation of the distributions for 5.75 MeV α-particles. The distribution w(E) on which the calculations are based is shown in Fig. 4. This spectrum is a combination of the classical formula for higher energies and of the experimental results of Rauth and Simpson [15] for very low energies. The shape of the spectrum at intermediate energies has been chosen in such a way that the right value for the stopping power results. Though this is only an

estimate of w(E), it is certainly much more realistic than the idealized relation $w(E) \sim 1/E^2$. While the theoretical value of δ_2 is 400 eV (see formula (13)), a value of 429 eV is obtained for the corrected spectrum. The mean energy δ_1 transferred in a primary collision is 62.5 eV (the theoretical value for δ_1 is only a few eV and has certainly no real meaning). The curves in Fig. 2 correspond to mean energy losses of 63 eV, 250 eV, 1 keV, 4 keV and 16 keV. The value of the parameter κ of straggling theory is 0.0026, 0.011, 0.042, 0.17, and 0.67.

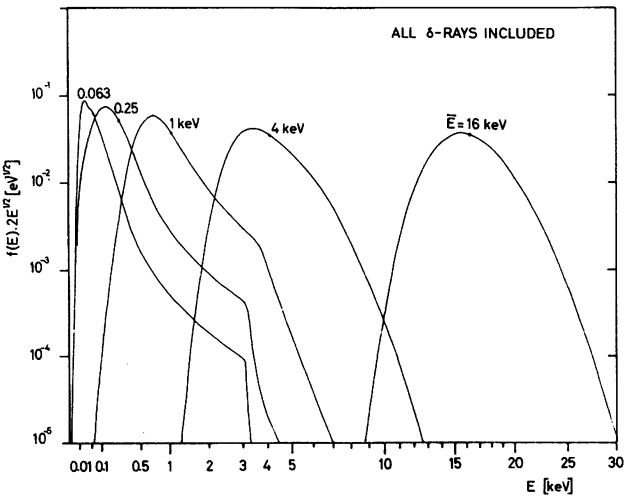

FIG. 2. Distributions of energy loss for 5.75 MeV α-particles in water. The corresponding distances in water are 8Å, 30 Å, 120 Å, 500 Å and 0.19 μm

The curves deviate markedly from the Vavilov distributions for small numbers of primary collisions. For higher mean energies they nearly agree with the Vavilov distributions, especially if the latter are corrected for glancing collisions (see Rossi and Rosenzweig [16]).

The values of the track length in water which correspond to the distributions in Fig. 2 are 0.0008 μm, 0.003 μm, 0.012 μm, 0.05 μm, and 0.19 μm. Since the maximum range of δ-rays for a 5.75-MeV α-particle is 0.3 μm, the distributions have to be corrected for the escape of δ-rays. Only with this correction can they be used as the basis for the calculation of local energy density spectra in very small volumes. Figure 3 serves to show the effect of a cut-off at high δ-ray energies. For these curves the spectrum w(E) (Fig. 4) is extended only up to 1 keV.

A comparison of both figures shows that, for small mean energy losses (small κ), the curves coincide over a wide range. Only the tails are different. However, for larger mean energy losses the difference becomes very significant. The effective κ of the curves in Fig. 3 is approximately three times greater than the κ value of the curves in Fig. 2. In other words, the variance of the curves is very strongly affected by

the cut-off. The value of δ_2 is decreased to 179 eV. While for low κ only the tail of the curves is altered, the change becomes decisive as soon as the curves approach the Gaussian shape. This is, of course, specifically important for radiobiological applications in the case of fast, densely ionizing particles.

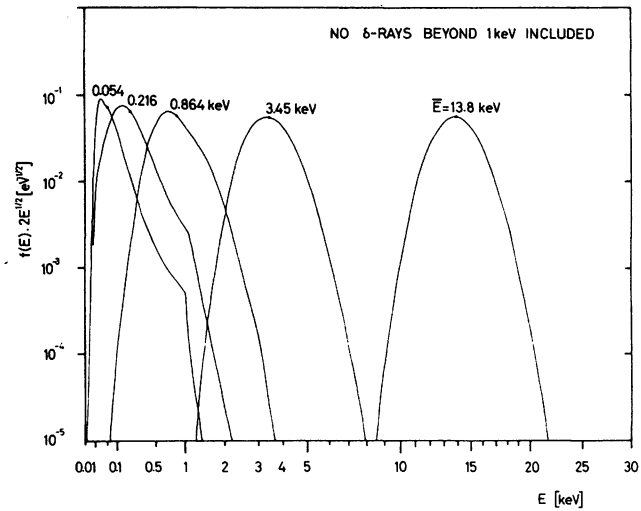

FIG.3. Distributions of energy loss for 5.75 MeV α-particles in water. The curves correspond to the distributions from Fig.2, but all δ-rays with energies beyond 1 keV are excluded

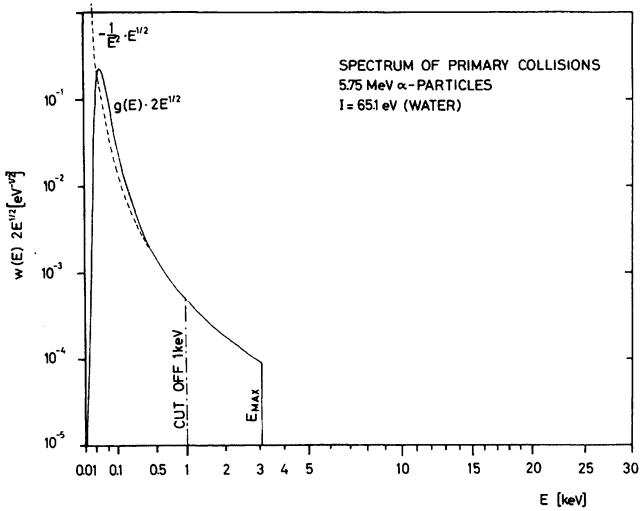

FIG.4. The spectrum w(E) of primary collisions has been used for the calculations. The dotted line indicates the $1/E^2$-spectrum which is used for the classical theories

The present example is rather crude. In practical cases one has to calculate escape probabilities for the different δ-rays. Then, instead of applying a simple cut-off, one may change the high-energy end of the distribution w(E) in a more appropriate manner.

To arrive at a better knowledge of the distribution w(E) further experiments, especially with rather slow particles, and with thin solid state detectors, with foils, with gas gaps, or with wall-less proportional counters, will be useful. In this connection the results of straggling experiments, straggling theory, and microdosimetry will be of great mutual interest.

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