taking  $v = 2 \times 10^4$  cm/sec, we obtain

$$\delta = 2.8\mu^{\frac{1}{3}} (\Delta \bar{\nu})^{\frac{2}{3}}.$$
 (27A)

Now the relevant wave-number separations in our case are (in the subscript notation of the excited states employed in the text)

$$\Delta \bar{\nu}_{1,2a} = 0.47 \text{ cm}^{-1},$$
  

$$\Delta \bar{\nu}_{1,2b} = 0.26 \text{ cm}^{-1}.$$
(28A)

PHYSICAL REVIEW

The values of the  $\delta$ 's corresponding to (28A) and to the two different values of  $|\mu|$  are given in Table I.

From these numbers and from the remarks subsequent to Eq. (25A) we arrive at the conclusion stated in the text subsequent to Eq. (11); namely, the energylevel discrepancy is too small to cause any order-ofmagnitude diminution of the cross section from its resonance value.

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# The Response of Anthracene Scintillation Crystals to High Energy $\mu$ -Mesons<sup>\*,†</sup>

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The response of an anthracene scintillation counter to high energy charged particles which lose only a small fraction of their energy in traversing the crystal was determined, using  $\mu$ -mesons in the cosmic radiation at sea level with energies from 29 Mev to greater than 1 Bev. The light output was found to have a sizeable fluctuation for mesons of the same initial energy, due to ionization loss straggling. The scintillation efficiency of the phosphor was found to decrease for increasing specific ionization, in agreement with the work of others on electrons and protons. The response of the crystal showed no rise within 2 percent for relativistic meson energies, which agrees with calculations of the density effect reduction in ionization loss for anthracene.

'HE light" output of scintillation crystals has been shown to be approximately proportional to the total ionization energy loss for low energy particles which spend their entire range in the crystal.1 Such proportionality between energy loss and light output would also be expected to be true for high energy charged particles which pass completely through the crystal and lose only a small fraction of their total energy by ionization in the crystal. It was the purpose of this work to find the light output of an anthracene crystal as a function of the energy of the traversing particle. Anthracene was used for this investigation because it has the largest light output of the known organic phosphors. µ-mesons from the cosmic radiation at sea level provided a good source of particles for such an experiment, because a wide range of energies is available and absorption by radiation losses and by nuclear collisions is negligibly small. The results which would be found for other charged particles should be the same as for  $\mu$ -mesons, except for a simple change of scale.

### I. THEORY

For the case of a charged particle traversing a thin absorber, a large fluctuation in the ionization energy

loss is to be expected. This "straggling" has been calculated by Williams<sup>2</sup> and, later, more accurately by Landau<sup>3</sup> and Symon.<sup>4</sup> The straggling is essentially caused by the fact that large energy transfers to single electrons can occasionally occur. These electrons, which are seen as "&-rays" in nuclear emulsions or as "knock-on electrons" in cosmic-ray work, lose their energy in the crystal in most cases; hence, the light output is increased. For high energy particles, where

 $W \gg \xi$ .

with

$$W \cong 2mc^2\beta^2/(1-\beta^2) \text{ (mesons and protons)},$$
  
$$\xi = 2\pi ne^4 x/mc^2\beta^2,$$
(2)

the energy loss distribution approaches a form which can be expressed in terms of a universal function. Here n is the electron density, m is the electron mass, e is the electronic charge, c is the velocity of light,  $\beta$  is v/c for the incident particle, and x is the absorber thickness in cm. W is the maximum energy loss possible in a single collision, and  $\xi$  is a parameter with the dimensions of energy which is a measure of the thickness of the absorber. If the probability of an energy loss between  $\epsilon$  and  $\epsilon + d\epsilon$  is  $P(\xi, \epsilon)d\epsilon$  in an absorber with a thickness parameter  $\xi$ , then it was shown by Landau<sup>5</sup> that

$$P(\xi, \epsilon) = \frac{1}{\xi} \phi \left( \frac{\epsilon - \epsilon_{\text{prob}}(\xi)}{\xi} \right).$$
(3)

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<sup>\*</sup> Assisted by the joint program of the ONR and AEC.

<sup>Assisted by the joint program of the UNK and AEC.
† Preliminary results of this investigation were reported in
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<sup>1</sup> W. H. Jordan and P. R. Bell, Nucleonics 5, 30 (1949); R.
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<sup>&</sup>lt;sup>2</sup> E. J. Williams, Proc. Roy. Soc. (London) 125, 420 (1929).
<sup>3</sup> L. Landau, J. Phys. (U.S.S.R.) 8, 201 (1944).
<sup>4</sup> K. R. Symon, Harvard University thesis (1948).
<sup>5</sup> See reference 3, Eq. (18).

The shape of  $\phi$  is illustrated by the smooth curve in Fig. 4(c). This distribution function has the property that an average of several energy loss measurements is no more accurate than a single measurement. For this reason, one must not deal with average energy losses, which are given in the usual energy loss formulas, but with the most probable energy loss, which is that loss for which the curve of frequency vs energy loss is a maximum.

The most probable loss is always less than the average loss, but for low energy particles, or large thicknesses of absorber, where

$$W \ll \xi$$
, (4)

the two become practically equal. Thus, at low energies or large absorber thickness, the usual formulas give the most probable loss directly. For a particle with kinetic energy, E, which is less than about  $Mc^2$ , we may use the Bethe-Bloch formula<sup>6</sup> for the average loss

$$\bar{\epsilon} = \xi \left[ \ln \frac{2mv^2 W}{I^2 (1-\beta^2)} - 2\beta^2 \right], \tag{5}$$

where I is the average ionization potential of the absorber. At high energies or small thickness, where condition (1) holds, one simply replaces the quantity corresponding to W in the logarithm by  $\xi \exp(0.37 + \beta^2)$ . Using formula (5) for the energy loss, we obtain<sup>7</sup>

$$\epsilon_{\text{prob}} = \xi \left[ \ln \frac{2.90 m v^2 \xi}{I^2 (1 - \beta^2)} - \beta^2 \right]. \tag{6}$$

Formulas (5) and (6) are not correct in the relativistic region because of the density effect, which was first calculated by Fermi<sup>8</sup> and later extended by Halpern and Hall,9 Wick,10 and Schönberg.11 Fermi calculated the energy loss of a charged particle in a dispersive medium whose electrons had a single characteristic frequency. He showed that the loss at extreme relativistic energies was dependent only upon the electron density of the medium. The later calculations<sup>9-11</sup> made use of multifrequency models, which gave slightly varying results in the transition region from nonrelativistic to extreme-relativistic particle velocities, but all agreed with Fermi's fundamental result beyond the transition region. If  $I_K$  is the binding energy of a K-electron in the element with the highest atomic number in the absorber, and if

$$\frac{\beta}{(1-\beta^2)^{\frac{1}{2}}} \gg \frac{I_K}{(ne^2h^2/\pi m)^{\frac{1}{2}}},\tag{7}$$

<sup>6</sup> See B. Rossi and K. Greisen, Revs. Modern Phys. 13, 247 (1941) Eq. (1.11) and for further references.

- <sup>7</sup> See reference 3, Eq. (15).
   <sup>8</sup> E. Fermi, Phys. Rev. 57, 485 (1940).
   <sup>9</sup> O. Halpern and H. Hall, Phys. Rev. 57, 459 (1940); 73, 477 (1948).
- <sup>10</sup> G. C. Wick, Nuovo cimento (9), 1, 302 (1943).
   <sup>11</sup> M. Schönberg, Nuovo cimento 8, No. 3 (1951).



FIG. 1. Arrangement A for nonrelativistic meson energies.

then the most probable energy loss in an absorber satisfying Eq. (1) is given  $by^{12}$ 

$$\epsilon_{\rm prob} = \xi \ln \left[ \frac{2.9mv^2 \xi}{(ne^2 h^2 / \pi m)} \right]. \tag{8}$$

For anthracene, condition (7) is equivalent to requiring that the energy of a  $\mu$ -meson must be much greater than 1.7 Bev to make Eq. (8) valid.

## **II. EXPERIMENTAL ARRANGEMENT**

The experiments to find the light output of the scintillation crystal as a function of meson energy and to check the validity of the theory outlined before were divided into two parts: nonrelativistic and relativistic meson energies.

The arrangement for the nonrelativistic energies (arrangement A) is shown in Fig. 1. Lead absorbers were used to select three ranges of meson energies, which were (a) 29 to 48 Mev, (b) 48 to 170 Mev, and (c) greater than 170 Mev.

A lead absorber above the apparatus caused  $\mu$ -mesons to lose about 600 Mev before entering the scintillation crystal, which increased the number of low energy mesons passing through the crystal, since the peak in the meson spectrum at sea level is in the neighborhood of 600 Mev. Coincidence ABDE-I defined a narrow

<sup>&</sup>lt;sup>12</sup> Equation (8) for the most probable loss was found by subtracting from Eq. (6) a correction term which is given, e.g., by Eq. (39) of reference (9), or by Eq. (22) of reference 10.



FIG. 2. Block diagram of recording circuits.

beam of particles which must traverse the anthracene scintillation crystal, C. The crystal was viewed by two 5819 photomultiplier tubes, one at each end of the cylindrical crystal (3-cm diam and 3 cm long). The remaining surface of the crystal was covered with aluminum foil to improve the light collection. Geiger counters  $F_1$ ,  $F_2$ , G, and H covered the solid angle defined by *ABDE*. Counters I were for the detection of side showers, which were in anticoincidence with *ABDE*.

A fourfold coincidence of the crossed pairs of counters AB and DE with an anticoincidence of Iresulted in a master pulse which initiated the sweep of an oscilloscope. The pulses from the two photomultiplier tubes were delayed and clipped to a width of 1.5  $\mu$ sec by the use of delay line. One pulse was delayed by 4  $\mu$ sec and the other by 7  $\mu$ sec, so that they appeared independently at separate positions on the oscilloscope trace. Pulses from G-M counters  $F_1$ ,  $F_2$ , G, and H were delayed with multivibrators by 15, 20, 25, and 30 µsec, respectively, and also appeared independently on the trace. The oscilloscope trace was photographically recorded by a camera which was equipped so that the master pulse automatically moved the film ahead for another event (see Fig. 2). This system made it possible to obtain the data for all three energy ranges during the same run of the equipment so that slow time variations in the apparatus could not affect the results in comparing the data for the various energy ranges.

Ordinarily, if only a single particle traversed the telescope, either  $F_1$  or  $F_2$ , but not both, would fire. The presence of pulses from both  $F_1$  and  $F_2$  served as an additional indicator of showers and knock-on electrons, which were eliminated from the analysis. Only events in which the master coincidence ABDE-I was accompanied by pulses from counters (a)  $(F_1 \text{ or } F_2)-(GH)$ , (b)  $(F_1 \text{ or } F_2)+G-H$ , or (c)  $(F_1 \text{ or } F_2)+G+H$  were

used, as these represent the three energy ranges for  $\mu$ -mesons. Very few single electrons should have been present, as it was almost impossible for them to get through the large thickness of lead above the apparatus.

The arrangement for relativistic meson energies (arrangement B) is shown in Fig. 3. Lead absorbers determined three energy bands for  $\mu$ -mesons from (d) 190 to 460 Mev, (e) 460 to 960 Mev, and (f) higher than 960 Mev. Coincidence ABCD defined a beam of mesons which must pass through two separate, identical crystals, 1 and 2 (3-cm diam and 3 cm long). Each crystal was viewed at one end by a 5819 photomultiplier tube, which made possible two completely independent measurements of the ionization loss of a particular meson. The surfaces of the crystals, except for the ends used for viewing, were again covered with aluminum foil. Trays E, F, and G covered the solid angle defined by ABCD. The recording system was identical to that in arrangement A, with the master coincidence ABCD, and the delayed pulses from photomultipliers 1 and 2 and from counters E, F, G, and H.

Counters H detected events accompanied by side



FIG. 3. Arrangement B for relativistic meson energies.



FIG. 4. Pulse-height histograms for energy ranges (a) through (f). The smooth curves are the theoretically expected curves due to energy loss fluctuations.

showers, which were eliminated from the analysis. Only those events in which the master coincidence ABCDwas accompanied by pulses from counters (d) E-(FG), (b) EF-G, or (f) EFG were used, as these represent the three energy ranges for  $\mu$ -mesons. Single electrons at sea level should have been stopped in the first 12.7 cm of lead, and therefore did not reach tray E.

Although energy loss fluctuations were important in the scintillation crystal, they resulted in only a 3 percent fluctuation in the range required to bring a meson to rest for the energies used in these experiments. Such a small variation could be completely neglected, since the ionization loss in the scintillation crystal was a slowly varying function of the energy. If high energy mesons were multiply scattered by the lead absorbers out of the solid angle covered by the G-M counter trays, then they would be counted as low energy particles and would affect the pulse-height distributions found for the



low energy ranges. It was estimated from calculations of the multiple scattering and from the counting rates obtained that no range contained more than 25 percent of extraneous high energy particles, which may be tolerated for the purposes of these experiments.

It was assumed for the analysis of most of the data that the heights of the pulses from the photomultiplier tubes were proportional to the energies lost in the crystals. This assumption was based upon the evidence (see Table III) that the proportionality factor between light output and energy loss was a slowly varying function of the specific ionization, which was close to minimum for the  $\mu$ -mesons and over most of the range of the electrons used for calibration.

The pulse-height scale with arrangement B was calibrated in units of energy loss (Mev) by comparison with the peak of the Compton electron distribution from the 2.62-Mev gamma-line of ThC". The pulse-

Average energy		Pulse height (Mev)	Energy loss from theory (Mev)	Scintillation efficiency
(a)	38 Mev	$9.7 \pm 0.7$	11.6	$0.84 \pm 0.06$
(b)	109 Mev	$7.2 \pm 0.3$	7.4	$0.97 \pm 0.03$

TABLE I. The most probable pulse heights for energy ranges (a) and (b).

height scale of arrangement A was normalized to arrangement B by the comparison of the distribution of pulse heights for E > 170 Mev (range c) in A with the distribution of E > 190 Mev (ranges d, e, and f) in B. This calibration made possible absolute comparisons with the theory to an estimated accuracy of about  $\pm 5$ percent. However, relative comparisons could be made to within about  $\pm 2$  percent for ranges d, e, and f.

### **III. RESULTS**

The frequency vs pulse-height histograms for energy ranges (a), (b), and (c) are shown in Fig. 4(a), (b), (c). Although only a few events were obtained in range (a), a peak at 9.6 Mev is distinctly visible. The pulses below 8 Mev and above 12 Mev were probably due to knock-on electrons and small air showers, which might cause one or two electrons at minimum ionization to traverse the crystal. The distribution for range (b) has its peak at about 7 Mev, which is significantly higher than the peak at 6 Mev for range (c). Range (c) corresponds to mesons which, as will be shown below, all lose the minimum possible energy. The pulse-height distribution for range (b) is somewhat broader than that for range (c) because of the fact that there was a continuous decrease in energy loss throughout range (b). The energy loss distribution curve calculated by Landau, which has been normalized for equal areas under the histogram and curve, is seen to be in good agreement with the experimental distribution of losses for minimum ionization mesons [Fig. 4(c)]. The positions of the peaks in the pulse height distributions, together with the theoretical values, are given in Table I.

Three of the six distributions found for ranges (d), (e), and (f) of experiment B are shown in Fig. 4(d), (e), (f). They are all seen to be in agreement with the theoretically expected distribution. Corrections due to variations in path length made possible by the cylindrical shape of the crystals have been made to the theoretical curves. The positions of the peaks of the distribution curves are given in Table II. It can be seen

TABLE II. The most probable pulse heights for energy ranges (d), (e), and (f).

Average energy		Counter 1	Counter 2	Average of 1 and 2
(d)	325 Mev	$6.07 \pm 0.15$	$6.14 \pm 0.15$	$6.11 \pm 0.10$
(e)	710	$6.19 \pm 0.12$	$6.17 \pm 0.12$	$6.18 \pm 0.08$
(f)	2700	$6.15 \pm 0.07$	$6.15 \pm 0.07$	$6.15 \pm 0.05$

that the most probable pulse heights are all equal within experimental error.

The fact that in arrangement A two photomultiplier tubes viewed one crystal, whereas in B they viewed two entirely separate crystals, allows us to definitely establish the source of the distribution in pulse heights which was found. It was found in experiment A that a large pulse from one phototube was invariably accompanied by a large pulse of practically the same height from the other. Figure 5A shows the most probable height of pulse 1 for a given height of pulse 2. In the case of two photomultiplier tubes viewing the same crystal, the two pulses were highly correlated. Except for a small remaining fluctuation of the order of 5 percent, light collection and the photoelectron multiplying process are ruled out as sources of the pulse-height distributions which were found.

In experiment B, the height of pulse 1 was found to be almost completely unrelated to the height of pulse 2,



FIG. 5. A. The dependence of the height of the pulse from photomultiplier No. 1 upon the height from No. 2 when both are viewing the same crystal. B. Conditions similar to A, except that photomultipliers are viewing separate crystals.

as can be seen in Fig. 5B. Since both pulses were the result of the same high energy meson, this indicates that the fluctuation was primarily caused by the energy loss mechanism, and could not be due to a hypothetical large variation in the probable ionization with the meson energy. This is just what one would expect from the theory, as the probable ionizations of all high energy mesons should have been equal, and large fluctuations should have been present in the ionization loss in traversing thin absorbers. The slight dependence of the height of pulse 1 upon pulse 2 at small pulse-heights was probably caused by mesons which traverse unusually short paths in the cylindrical crystals. A particle which was close enough to the side of one crystal so as to appreciably shorten the path length must also have been close to the side in the other crystal, because of the G-M counters being some distance away from the crystals; hence a short path in one crystal implied a short one in the other, also.

A reduction in the scintillation efficiency of anthracene with increasing specific ionization is evident from an inspection of Table I, since the ionization loss theory must be regarded as well-verified for energies below the minimum ionization region. Table III compares these efficiencies with values found for electrons.<sup>13</sup> These results seem to indicate that the nonlinearity of the response of the scintillator is caused by a saturation effect which is a function only of the specific ionization and is independent of the type of particle.

The values found for the most probable energy losses for energy ranges (a), (b), (d), (e), and (f) are compared with the theoretically expected curve in Fig. 6. The points at (a) 38 and (b) 109 Mev were corrected for the loss in efficiency of the crystal, using the values for electrons listed in Table III, since the efficiencies must be taken from an independent source. The remaining three points are the averages from Table II. No scintillator efficiency correction was required for these



FIG. 6. The most probable energy loss in the anthracene crystal as a function of meson energy. Solid curve: theory with density effect correction. Dotted curve: Theory without density effect correction.

points, since they were at minimum ionization. The density effect correction for anthracene was estimated to be a function similar in shape to those for carbon and water as found by Halpern and Hall<sup>9</sup> and Wick.<sup>10</sup> At nonrelativistic energies the correction was assumed to

TABLE III. Scintillation efficiency of anthracene.

Specific ionization	Scintillation efficiency		
(Min ion =1)	Mesons	Electrons	
1.00	1.00 (definition)	1.00	
1.23	$0.97 \pm 0.03$	0.98	
1.93	$0.84 \pm 0.06$	0.89	

approach zero, and at extreme relativistic energies it was made so as to give a result in agreement with Eq. (8). The uncertainty of the ordinates of the theoretical curve with the density effect correction were estimated to be of the order of 2 or 3 percent. As a consequence of the low effective Z of anthracene, the calculated curve shows practically no relativistic rise in the probable ionization loss. Similar curves for higher Z absorbers generally would be expected to show some rise beyond the minimum before leveling off. The experimental points are seen to be in excellent agreement with the theoretical curve corrected for the density effect in anthracene, and indicate that between 300 and 3000 Mey there is no relativistic rise within 2 percent in the most probable ionization loss. The dashed curve calculated from the Bethe-Bloch formula (6) for the most probable loss without the density effect correction is seen to lie well outside the experimental points. These results appear to definitely establish the existence of the reduction in ionization loss due to the density effect.

### IV. CONCLUSIONS

The results of this work show that in dealing with scintillations caused by particles which lose only a small fraction of their energy in traversing the scintillator, one must necessarily have a sizeable fluctuation in light output because of straggling in the ionization energy loss. A reduction in the scintillation efficiency of anthracene with increasing specific ionization has been found for  $\mu$ -mesons which is noticeable (0.84±0.06) even when the specific ionization is only double minimum ionization. The results for relativistic energies show no relativistic rise in ionization loss in anthracene, which can be regarded as a complete verification of the existence of the density effect.

The authors wish to express their appreciation to Professor Marcel Schein for his constant interest and guidance in this work.

<sup>&</sup>lt;sup>13</sup> Data taken from graph in paper by Frey, Grim, Preston, and Gray, Phys. Rev. 82, 372 (1951), which was based on data in paper by J. I. Hopkins, Phys. Rev. 77, 406 (1950).