Ionization Energy Loss of Mesons in a Sodium Iodide Scintillation Crystal*

THEODORE BOWEN†
Department of Physics, University of Chicago, Chicago, Illinois
(Received June 21, 1954)

The response of a thallium-activated sodium iodide crystal to high-energy charged particles passing through the scintillator has been investigated. Pions and muons produced by the Chicago 450-Mev cyclotron have been used to cover energies ranging from 61 Mev (pions) to 245 Mev (muons). Sea-level cosmic-ray muons have been used to cover energies from 200 Mev to greater than 2 Bev. At each energy, the most probable energy loss is determined from the pulse-height distribution, making use of Po-Be 4.44-Mev γ rays for an energy calibration. The distributions from cyclotron data are found from the density variations on photographs of the superposition of many pulses appearing on an oscilloscope. Because the cosmic-ray counting rate is low, each pulse is individually photographed and measured. The results for energies below the occurrence of the minimum in ionization indicate good agreement with the probable energy loss as given by the Bethe-Bloch formula. Beyond the minimum the probable loss is in fair agreement with Sternheimer's calculations for sodium iodide, rising slightly more rapidly than predicted to the Fermi plateau.

INTRODUCTION

An exact understanding of energy losses by high energy charged particles has long been the object for much theoretical and experimental work. An accurate knowledge of the behavior of ionization energy loss not only helps us to select the best theoretical model for the phenomena, but also makes it possible to use ionization loss along the path of a particle as a quantitative tool in the carrying out of experiments on the properties of high-energy particles. The comparatively recent development of scintillation-counting techniques provides an attractive method of measuring the specific ionization of fast particles, and thus offers the possibility of making new and more accurate comparisons with ionization loss theory.

The first theory of ionization energy loss was developed by Bohr[1] along essentially classical lines. The theory was improved by the quantum mechanical calculations of Bethe and Bloch,[2] and the formula derived by them for the specific energy loss as a function of the velocity of the charged particles has come to be known as the Bethe-Bloch formula. By suitably adjusting the one parameter, the average ionization potential of the absorber, excellent agreement between theory and experiment can be obtained for nonrelativistic particle energies.[3] When the Bethe-Bloch formula is extrapolated to the region of relativistic energy losses, one is led to expect the specific energy loss to increase logarithmically with the energy, with no upper limit to the loss which can be reached.

Swann[4] first suggested that polarization of the medium by the fields surrounding the moving charged particle might reduce the energy loss to be expected at high energies. This polarization or density effect was first calculated quantitatively by Fermi,[5] who showed that for extreme relativistic particles the rate of energy loss depends only upon the electron density of the absorber. If, as in most experimental conditions, the occasional energy losses to single electrons larger than some arbitrary energy, T, are ignored, then the energy loss per unit of path length is expected to reach a constant plateau at extreme relativistic energies. Fermi also showed that part of the loss at relativistic energies was in the form of Čerenkov radiation.

The theory of the density effect was then extended by Halpern and Hall[6] and Wick[7] by introducing multifrequency models of a dispersive medium into the calculations. Further studies of the energy loss of relativistic particles have been made by A. Bohr, Schönberg, Messel and Ritson, Huybrechts and Schönberg, Sternheimer, Budini and Poiani, and Fowler and Jones.[8–11] From the earlier work[8] based on the Lorentz dispersion model it appeared that the relativistic rise in the energy loss escaped in the form of Čerenkov radiation, and hence, would not be expected to contribute to the ionization observed close to the path of the particle. However, it has been shown[10,11] that when the dispersion model of the Kramers-Kallmann-Mark theory based on x-ray work is used, very little energy is lost in the form of Čerenkov radiation, and the relativistic rise appears in the form of ionization close to the path of the particle. The

* Supported in part by a joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.
† U. S. Atomic Energy Commission Predoctoral Fellow during part of this work.
[1] N. Bohr, Phil. Mag. **25**, 10 (1913); **30**, 581 (1915).
[2] H. Bethe, Ann. Physik **5**, 325 (1930); Z. Physik **76**, 293 (1932); F. Bloch, Ann. Physik **16**, 285 (1933); Z. Physik **81**, 363 (1933).
[3] S. K. Allison and S. D. Warshaw, Revs. Modern Phys. **25**, 779 (1953).
[4] W. F. G. Swann, J. Franklin Inst. **226**, 598 (1938).
[5] E. Fermi, Phys. Rev. **56**, 1242 (1939); **57**, 485 (1940).
[6] O. Halpern and H. Hall, Phys. Rev. **57**, 459 (1940); **73**, 477 (1948).
[7] G. C. Wick, Ricerca sci. **11**, 273 (1940); **12**, 858 (1941); Nuovo cimento **1**, 302 (1943).
[8] A. Bohr, Kgl. Danske Videnskab. Selskab. Mat.-fys. Medd. **24**, No. 19 (1948); M. Schönberg, Nuovo cimento **8**, 159 (1951); H. Messel and D. Ritson, Phil. Mag. **41**, 1129 (1950).
[9] M. Huybrechts and M. Schönberg, Nuovo cimento **9**, 764 (1952).
[10] R. M. Sternheimer, Phys. Rev. **88**, 851 (1952); **91**, 256 (1953).
[11] P. Budini and G. Poiani, Nuovo cimento **9**, 199 (1952); P. Budini, Phys. Rev. **89**, 1147 (1953); G. N. Fowler and G. M. D. B. Jones, Proc. Phys. Soc. (London) **A66**, 597 (1953).

results of this work will be compared mainly with the calculations of Sternheimer,[10] since he has made explicit numerical computations for sodium iodide.

In order to make measurements of the rate of energy loss of a fast particle, it is generally necessary to determine the energy lost along a small, fixed length of its path. Unfortunately, this loss is subject to large fluctuations or straggling due to the occasional production of high-energy δ rays or knock-on electrons. The energy loss straggling in thin absorbers was first calculated by Williams[12] and later more accurately by Landau.[13] Symon[14] extended the theory to the intermediate case between a thin absorber which gives rise to a Landau distribution and a thick absorber which gives rise to a Gaussian distribution of energy losses. Blunck and Leisegang[15] have computed the effect of resonances in the energy loss process which cause a broadening of the Landau distribution for very thin absorbers where the loss is of the same order of magnitude as the atomic ionization potential of the absorber. Thus, in comparing energy loss theories with experiment, it is not only necessary to correct the average energy loss as computed from the Bethe-Bloch formula for the density effect, but also for the effect of straggling.

Recent measurements[16] of the ionization in gases using cloud chambers, proportional counters, inefficient counters, and ion chambers show a definite relativistic increase and appear to be in good agreement with the most recent theoretical calculations.[10,11] However, over the range of energies and gas pressures studied in most of this work, the energy loss is given by the Bethe-Bloch formula, so very little information has been gained in the region where the density effect becomes important.

Whittemore and Street,[17] by comparing the pulse distributions from an AgCl crystal counter for two meson-energy ranges, were among the first to find convincing experimental evidence of the density effect. The energy loss of 9.6- and 15.7-Mev electrons from a betatron in a number of solids has been measured using a magnetic analyzer by Goldwasser, Mills, and Hanson.[18] They find good agreement with the theory using the density effect correction, except for the heaviest elements. More recently, Goldwasser, Mills, and Rubillard[19] have directly observed the density effect by observing the energy loss in the same chemical compound in the gaseous and solid states and find good agreement with the theory. While these data check the position of the Fermi plateau, they do not yield information about the shape of the energy loss curve before reaching the plateau.

Measurements of grain density in photographic emulsions show a definite relativistic increase.[20] However, the various workers do not agree too well on the exact magnitude and energy dependence of the relativistic rise. Part of the discrepancy may be due to the inaccuracy of momentum measurements in the region where the relativistic rise takes place. In addition, as has been pointed out by Brown,[21] the observed grain densities may not always be proportional to the total energy loss because an energy threshold must be exceeded in order to cause a grain to develop. Therefore, it may be premature to draw definite conclusions from the agreement or lack of agreement of grain density measurements with theory.

The ionization energy loss of high-energy particles can be studied as a function of particle energy by means of the pulse height distributions produced in scintillation counters. This has been demonstrated using cosmic-ray muons by the work of Bowen and Roser,[22] Meshkovskii and Shebanov,[23] and Baskin and Winckler[24] on the organic scintillators anthracene, stilbene, and terphenyl in xylene, respectively. All find good agreement with the theory when the density effect is taken into account. Hudson and Hofstadter[25] find that the pulse height distribution in sodium iodide due to sea level cosmic-ray mesons is only consistent with the theory when a density effect is included. However, since the mesons were not separated into energy groups, they could not study the detailed behavior of the energy loss as a function of the meson energy.

On the basis of the work cited above, energy loss as a function of the particle energy seems well established in gases and in condensed media of low atomic number. However, more data on condensed media of relatively high atomic number are needed to establish the energy loss curve in the region of minimum ionization and

[12] E. J. Williams, Proc. Roy. Soc. (London) **125**, 420 (1929).
[13] L. Landau, J. Phys. (U.S.S.R.) **8**, 201 (1944).
[14] K. R. Symon, Harvard University thesis, 1948 (unpublished). Most of the results are published in *High Energy Particles* by B. Rossi (Prentice-Hall Inc., New York, 1952).
[15] O. Blunk and S. Leisegang, Z. Physik **128**, 500 (1950).
[16] M. H. Shamos and I. Hudes, Phys. Rev. **84**, 1056 (1951); Gosh, Jones, and Wilson, Proc. Phys. Soc. (London) **A65**, 68 (1952); R. S. Carter and W. L. Whittemore, Phys. Rev. **87**, 494 (1952); G. W. McClure, Phys. Rev. **87**, 680 (1952); Price, West, Becker, Chanson, Nageotte, and Treille, Proc. Phys. Soc. (London) **A66**, 167 (1953); Perry, Rathgeber, and Rouse, Proc. Phys. Soc. (London) **A66**, 541 (1953); R. H. Frost and C. E. Nielson, Phys. Rev. **91**, 864 (1953); J. E. Kupperian, Jr., and E. D. Palmatier Phys. Rev. **91**, 1186 (1953); H. R. Snodgrass, Phys. Rev. **92**, 1089 (1953).
[17] W. L. Whittemore and J. C. Street, Phys. Rev. **76**, 1786 (1949).
[18] Goldwasser, Mills, and Hanson, Phys. Rev. **88**, 1137 (1952); C. Warner, III, and F. Rohrlich, Phys. Rev. **93**, 406 (1954).
[19] Goldwasser, Mills, and Rubillard, Phys. Rev. **90**, 378 (1953).
[20] E. Pickup and L. Voyvodic, Phys. Rev. **80**, 89 (1950); I. B. McDiarmid, Phys. Rev. **84**, 851 (1951); A. H. Morrish, Phil. Mag. **43**, 533 (1952); Daniel, Davies, Mulvey, and Perkins, Phil. Mag. **43**, 753 (1952); L. Jauneau and F. Hug-Bousser, J. Phys. et radium **13**, 465 (1952); M. M. Shapiro and B. Stiller, Phys. Rev. **87**, 682 (1952); R. P. Michaelis and C. E. Violet, Phys. Rev. **90**, 723 (1953); A. H. Morrish, Phys. Rev. **91**, 423 (1953); J. R. Fleming and J. J. Lord, Phys. Rev. **92**, 511 (1953); B. Stiller and M. M. Shapiro, Phys. Rev. **92**, 735 (1953).
[21] L. M. Brown, Phys. Rev. **90**, 95 (1953).
[22] T. Bowen and F. X. Roser, Phys. Rev. **85**, 992 (1952).
[23] A. G. Meshkovskii and V. A. Shebanov, Doklady Akad. Nauk. S.S.S.R. **82**, 233 (1952).
[24] R. Baskin and J. R. Winckler, Phys. Rev. **92**, 464 (1953).
[25] A. Hudson and R. Hofstadter, Phys. Rev. **88**, 589 (1952).

beyond where the relativistic rise to the Fermi plateau takes place. An accurate knowledge of the rate of energy loss in this region should help to extend the energy region in which mass measurements and particle identifications can be made.

The work of Hofstadter[26,27] and others has shown that the thallium-activated sodium iodide scintillation crystal has many properties which make it ideally suited for investigating ionization energy loss. The high content of iodine ($Z=53$) gives it a relatively high mean atomic number, comparable to that of AgBr in emulsion; hence, the results from sodium iodide scintillators and emulsion should be similar. The high atomic number also makes possible precise absolute energy calibrations from photoelectrons and pairs produced by γ rays of known energy.[27] Taylor *et al.* [28] have shown that except for slow α particles, where the ionization is of the order of two hundred times minimum ionization, the light output of a sodium iodide crystal is proportional to the energy lost by the charged particle.

In order to investigate the specific energy loss in a scintillating material, particles of known energy must be allowed to pass through a fixed thickness of scintillator in which they lose only a small fraction of their initial energy. Because of straggling of the energy losses, the pulses from the scintillation counter will be distributed in pulse height. The distribution is very skew, with a long tail in the direction of large energy losses. The problem of determining the energy loss is, then, essentially the problem of determining the position of the pulse height distribution. Because of the nature of the distribution, the average pulse height is a very poor estimate of the position of the distribution, since it is subject to large fluctuations caused by occasional very large losses. It is much better to characterize the position of the distribution by the position of the maximum, i.e., by the most probable pulse height. Even though the width at half maximum of the distribution is greater than 20 percent, the most probable value can be obtained to any desired accuracy by obtaining a sufficient number of events. The experimental work was planned to give an accuracy of 1 percent or better in the determination of each probable energy loss.

If a complete investigation of energy loss in sodium iodide were to be undertaken, it seemed desirable to examine the shape of the ionization *versus* energy curve on both the nonrelativistic and relativistic side of the minimum. Mesons provide ideal particles for this energy range, since energy losses due to radiation are negligible. For nonrelativistic energies, mesons produced by the 450-Mev Chicago cyclotron are very suitable, since monoenergetic high intensity beams were available for pion energies up to 227 Mev. For higher energies, one can use muons up to 250 Mev, which is close to the energy for minimum ionization. For still higher energies in the relativistic range, one can use muons from the sea level cosmic radiation. The energy spectrum of muons at sea level has a broad maximum between 100 and 1000 Mev so that most of the mesons lie in the energy region of the relativistic increase in energy loss. The highest energy to which it is practical to go is determined partially by the decrease in meson intensities with higher energies and partially by the increasing difficulty of measuring high energies. The latter limitation was most important in this work where range was used, the largest range measurement corresponding to about 2 Bev. Because of the difference in experimental techniques, the study of energy losses is divided into two parts corresponding to work at energies up to the energy corresponding to minimum ionization using cyclotron-produced mesons and the work at relativistic energies using cosmic-ray mesons.

CYCLOTRON ARRANGEMENT

The arrangement using the 227-Mev π^- beam from the cyclotron is shown in Fig. 1. Protons of 450-Mev energy striking an internal beryllium target produce pions, which are first magnetically analyzed by the

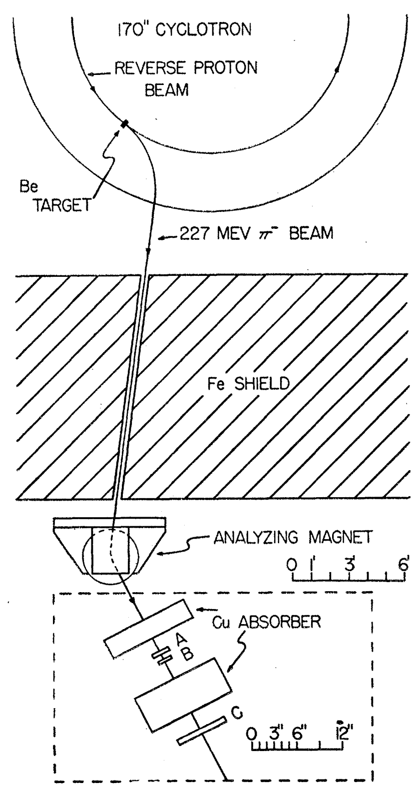

FIG. 1. Cyclotron arrangement for the 227-Mev π^- beam. The counter arrangement is shown in an enlarged scale.

[26] R. Hofstadter, Phys. Rev. **75**, 796 (1949).
[27] R. Hofstadter and J. A. McIntyre, Phys. Rev. **80**, 631 (1950).
[28] Taylor, Jentchke, Remley, Eby, and Kruger, Phys. Rev. **84**, 1034 (1951).

cyclotron's magnetic field. A channel through the shield wall is used which selects negative particles of 340 Mev/c momentum coming from the target. After passing through the shield wall, the particles are magnetically analyzed a second time by a deflection through 35°, giving a beam consisting of 97 percent 227-Mev negative pions and about 3 percent negative muons.[29] Since the muons mainly originate from the decay of pions close to the target and pass through two analyzing magnetic fields, they also have a momentum of 340 Mev/c, which corresponds to an energy of 250 Mev. Some data was also taken using the 122-Mev beam, the setup being similar except for the use of a different channel through the shield and different target and analyzing magnet settings.

The average energy of the 227-Mev beam has been determined by range measurements to within ± 3 Mev with a total spread in energies of less than 4 Mev. In the 122-Mev beam the energy is known to ± 2 Mev and the total spread in energies is less than 3 Mev. Other lower energies were obtained with each beam by introducing copper absorbers in front of counter A (Fig. 1). This introduces a small additional fluctuation in energy of the order of 3 percent due to energy loss straggling, as well as reducing the beam intensity by nuclear absorption and by nuclear and Coulomb scattering out of the beam.

A view of the counter arrangement is shown in Fig. 1 in an expanded scale. Scintillator A is the thallium-activated sodium iodide crystal[30] in which the energy loss was to be studied. It was $1.51 \times 3.47 \times 5.97$ cm^3 and was positioned so the 5819 photomultiplier tube viewed one end. The crystal was wrapped with an aluminum foil reflector and was hermetically sealed in an aluminum can with a Lucite window. The light collection was uniform to within ± 6 percent.

Counters B and C formed a coincidence telescope to select the mesons for which a measurement of the energy loss was to be made in the sodium iodide. Counter B was a $1.0 \times 2.5 \times 5.0$-cm^3 plastic scintillator[31] viewed at one end by a 5819 photomultiplier tube. The large counter C consisted of two $2 \times 6 \times 17$ cm^3 plastic scintillators, each viewed by a separate photomultiplier tube, and located side by side so that an effective area of 12×17 cm^2 was covered. The sensitive area was made large so that losses would be reduced due to small angle Coulomb and diffraction scattering in counters A and B and in any copper absorber which might be between B and C. With this arrangement, almost all the mesons selected by the coincidence BC have passed through the sodium iodide crystal A.

When it was desired to measure the energy loss of pions, no absorber was used between B and C. To measure the effect of the 250-Mev muons only, 126 g/cm^2 of copper absorber was inserted between counters B and C. The range of pions of 340-Mev/c momentum is 122 g/cm^2 Cu, whereas muons of the same momentum have a range of 144 g/cm^2 Cu. Since the total amount of material ahead of counter C was equivalent to 134 g/cm^2 Cu, the pions were unable to reach C and cause coincidences. However, the muons could still reach C and give coincidences. In this way it was possible to measure the energy losses due to muons.

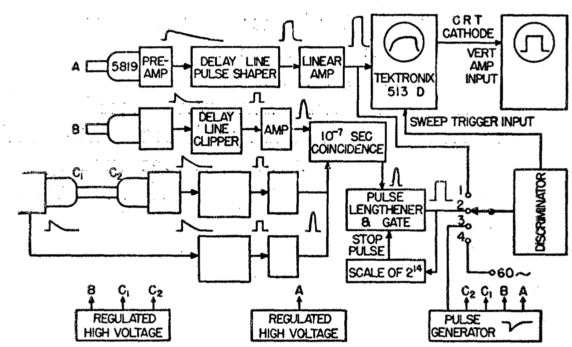

FIG. 2. Block diagram of the electronics for cyclotron produced mesons.

A block diagram of the electronics for cyclotron produced mesons is shown in Fig. 2. Counter B is in a 10^{-7}-second coincidence with the combined output of C_1 and C_2. This coincidence pulse, after passing through a gate circuit and discriminator, is used to trigger the sweep of a Tektronix 513D oscilloscope. The output of the sodium iodide scintillation counter A is shaped into a rectangular pulse 1.0 μsec wide by means of delay line. This pulse enters the vertical input of the oscilloscope. Since each meson which passes through B and C also has passed through A, each oscilloscope trace presents a pulse whose height is proportional to the energy lost by a meson in the sodium iodide A. A scale of 2^{14} circuit is also operated by the coincidence pulses. This circuit closes the gate circuit after $2^{14} = 16\,384$ events, thus stopping the recording until manually restarted. The input of the discriminator which triggers the oscilloscope sweep may be switched to other trigger sources for the purpose of recording the distribution due to the calibration γ-ray source (position 1), observing artificial test pulses from a mercury switch pulse generator (position 3), and recording a zero reference base line (position 4).

In the 227-Mev pion beam, typical counting rates were 9600 counts per minute for 227-Mev pions, 4100 counts per minute for pions slowed down to 167 Mev, 2100 counts per minute for pions slowed down to 121 Mev, and 360 counts per minute for 250-Mev muons. Although much higher rates were possible in the 122-Mev pion beam, the beam intensity was adjusted for similar rates to avoid any possibility of pulse pileup. When the duty cycle of the cyclotron is taken into account, the average spacing τ between counter pulses

[29] Anderson, Fermi, Martin, and Nagle, Phys. Rev. **91**, 155 (1953); M. Glicksman, Phys. Rev. **95**, 1045 (1954).

[30] Obtained from Harshaw Chemical Company, Cleveland, Ohio.

[31] C. N. Chou, Phys. Rev. **87**, 903 (1952). The author wishes to thank Dr. Chou for kindly supplying the scintillating plastic.

can be computed. At the highest intensity used, τ was 78 μsec, which was much greater than the 1-μsec. width of the pulses from the sodium iodide.

The pulse-height distributions were recorded by a 35-mm camera containing Linagraph Ortho film which viewed the oscilloscope screen. The lens opening and oscilloscope spot intensity were so adjusted that a single trace did not produce a noticeable blackening of the film, but an exposure to 2^{14} traces resulted in a density distribution on the film which was related to the frequency distribution of pulse heights. It was not necessary to know the exact relationship between density on the film and frequency since it was only necessary to find the most probable pulse height, which corresponds to the location of the region of greatest optical density on the film.

A calibration of the scale of pulse heights in Mev of energy loss was made possible by recording the pulse height distribution produced by a 2.5-millicurie polonium-beryllium neutron source. The main γ-ray produced by this source is one of 4.44 Mev from the decay of the first excited state of C^{12}.[32] This γ-ray produces a pair peak at 3.42 Mev which is used for energy calibration. Comparison of the pulse height of this pair peak with the 0.662-Mev Cs^{137} photopeak showed the response to be linear.

Before and after making each exposure to a pulse-height distribution, a zero reference line was recorded by exposing to about 300 blank traces. This was done at the beginning and end of each exposure to provide a check against possible drift of the oscilloscope image, since some of the exposures lasted longer than 30 minutes.

To eliminate the possibility of a slow drift of the sensitivity of the equipment while the measurements were being carried out, the distribution from pions of the full beam energy was repeated between each exposure to a different energy and between each γ-ray calibration. As the light intensity required for optimum exposure was rather critical, three exposures at each energy were taken differing by factors of two in light intensity. To obtain reproducible oscilloscope beam intensities, a second oscilloscope was used to observe the pulse across a small cathode resistor caused by the beam current.

In order to make more accurate estimates of the most probable pulse height, microphotometer tracings of the density distributions were made. The linearity between pulse height at the output of the photomultiplier and the measurement on the microphotometer tracing was checked by introducing artificial pulses of known amplitude ratios and was found to be within 1 percent over the region used.

COSMIC-RAY ARRANGEMENT

An apparatus was designed which would select cosmic-ray muons passing through a small area and solid angle where scintillation materials could be placed. In order to study the dependence upon meson energy of the scintillation responses, provision was made for classifying the selected mesons into four range intervals. Means were also provided to eliminate events due to electrons and showers. For each event, the responses of up to four separate scintillators could be simultaneously recorded. By providing an arrangement where the data over all the energy ranges for several scintillators could be collected simultaneously, not only was a large saving in operating time made possible, but accurate comparisons of the results from different energies and scintillators were made possible which are not subject to the errors of calibrations and slow drifts with time. The design follows very closely that of Bowen and Roser.[22]

The arrangement of counters and absorbers is shown in Fig. 3. Charged particles are selected by the side-by-side telescopes AB_1C_1D and AB_2C_2D. Either of these coincidences provides a master pulse which allows the recording of the pulse heights from the four scintillation counters and the recording of whether or

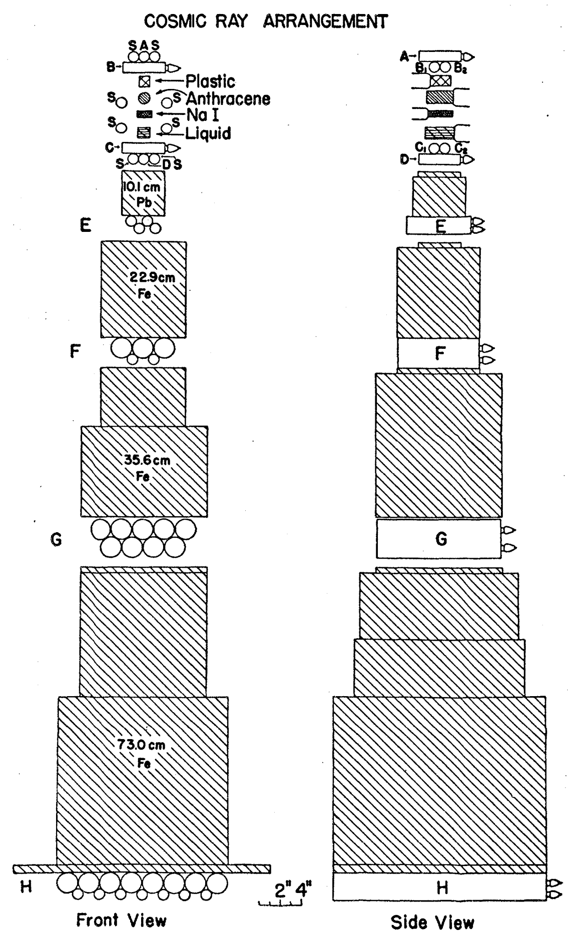

FIG. 3. Arrangement of counters and absorbers for cosmic-ray mesons. The dimension shown on each absorber refers to its vertical thickness.

[32] R. G. Thomas and T. Lauritsen, Phys. Rev. 88, 969 (1952).

not GM trays E, F, G, and H and shower counters S are in coincidence with the master pulse. The E, F, G, and H trays determine to which range interval the meson belongs. The S counters prove highly efficient in detecting when two or more particles are present, as judged from the observation that almost all anomalously large or small scintillation counter pulses are accompanied by S.

A block diagram of the electronics is shown in Fig. 4. A 10^{-5}-sec coincidence of AB_1C_1D or AB_2C_2D triggers the camera drive and dead-time circuit. If 12 seconds or more have elapsed since the preceding event so that the camera is ready, then a pulse is promptly sent on to trigger the sweep of the Tektronix 514D oscilloscope. The pulses from the scintillation counters are delay-line-shaped to a width of 1.5 μsec; they are then delayed by 0, 3, 6, and 9 μsec and mixed together. After a further 4-μsec delay, the group of pulses enters the vertical amplifier of the oscilloscope. In this way, each pulse is individually displayed. Pulses delayed 48, 42, 34, 27, and 20 μsec from counter trays S, E, F, G, and H, respectively, also appear on the oscilloscope sweep. These delayed pulses are obtained from the trailing edges of pulses from one shot multivibrators. Delayed pulses are also used to indicate which of the two possible master coincidences triggered the sweep.

The recording of the oscilloscope traces was made on Linagraph Ortho 35-mm film. Each time the oscilloscope sweep was triggered, the camera drive circuit started the camera motor to move the film in position for the next event. At the same time a mechanical register was also operated whose numbers could be correlated with the events on the film. Auxiliary feedback-type circuits had to be added to the oscilloscope to improve the stability of the vertical positioning and spot intensity, making the recordings of more uniformly good quality.

The four scintillators used were from top to bottom, anthracene in polystyrene plastic,[31] an anthracene crystal, a thallium-activated sodium iodide crystal, and terphenyl plus diphenylhexatriene in phenylcyclohexane liquid.[33] The final results from the three organic scintillators will be presented in a later publication. The sodium iodide crystal was the same one used with cyclotron-produced mesons with dimensions $1.51\times3.47\times5.97$ cm.[3] The crystal was viewed from one end as in the cyclotron arrangement by an RCA 5819 photomultiplier tube. Since the mesons selected by telescopes AB_1C_1D and AB_2C_2D passed through the crystal at different average distances from the photocathode, some information about the uniformity of the light collection efficiency could be gained.

The events could be classified into four meson energy ranges corresponding to the master coincidence being accompanied by E, EF, EFG, or EFGH. It was also required that there be no pulse from counters S and

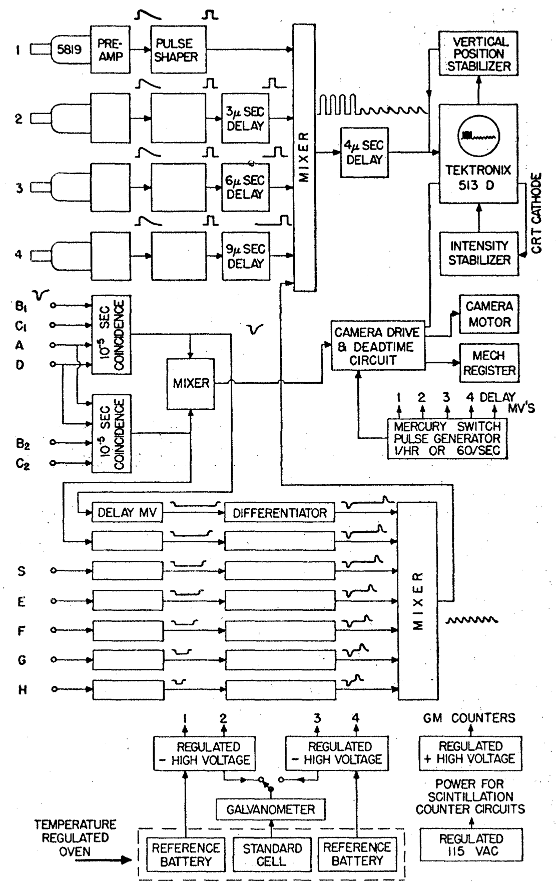

FIG. 4. Block diagram of the electronics for cosmic-ray mesons.

that all four scintillator pulses be present and at least one-half the most probable height. From an analysis of events which were otherwise acceptable, but which had one tray missing, such as FGH, or EFH, the efficiency of counter trays E, F, and G were found to be greater than 99 percent. Tray H could not be checked in the same way, but a comparison of single counting rates shows that its efficiency should be about 99 percent.

The energy spectrum of mesons selected by a particular coincidence is determined not only by the energy loss of the mesons in the absorbers between counter trays, but also by the amount of scattering which the mesons suffer. The first absorber is 10.1 cm of lead. Although mesons which have more than 193 Mev at the center of the sodium iodide crystal can pass through this lead, most electrons found at sea level will be stopped. The remaining absorbers, however, consist of iron, in which, for equal thicknesses, the rate of ionization energy loss is only about 9 percent less than that for lead, yet the rms multiple Coulomb scattering is less by a factor of about 1.88 because of the lower atomic number of iron. It is important in this application to reduce the multiple Coulomb scattering to as low a

[33] H. Kallmann and M. Furst, Nucleonics 8, No. 3, 32 (1951).

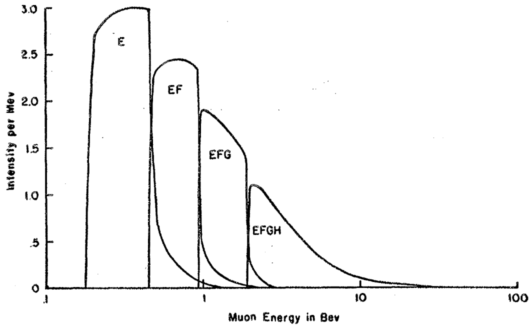

FIG. 5. Calculated energy spectra for E, EF, EFG, and EFGH events.

level as possible, since a high energy meson which is scattered out of an absorber will be counted as belonging to a lower energy interval.

The actual energy spectrum of mesons selected by each range interval has been computed. This was done by numerical computations which took account of energy loss in the absorbers and the probability of a muon being scattered out by multiple Coulomb scattering. The distributions of lateral deflections due to multiple scattering were approximated by Gaussian curves. The curve given by Rossi[34] was assumed for the energy spectrum of muons entering the apparatus. The resulting spectra for the four range intervals are shown in Fig. 5. By finding the area under the curves of Fig. 5, the results of these calculations can be used to predict the counting rates expected in each range interval. Table I gives a comparison between computed and actual counting rates. The agreement appears satisfactory in view of the approximations made in the calculations and the uncertainties in the meson spectrum.

Since the cosmic-ray apparatus had to be operated over a period of more than three months, it was necessary to provide for a high degree of stability. It was especially important to provide a very stable high voltage to the photomultiplier tubes, since a change of 1 percent in the high voltage would result in a 10 percent change in photomultiplier gain. Consequently, the electronically regulated supplies for the photomultipliers used batteries as voltage references. These reference batteries were enclosed along with a standard cell in an oven whose temperature was regulated at $(30.5\pm0.5)\,^{\circ}$C. The high voltage from either supply could be checked against the standard cell by means of a voltage divider using precision wire wound resistances and a null indicating galvanometer. With the above arrangement, the high voltage supplied to the photomultiplier tubes was kept well within ±0.1 percent.

Outputs from a mercury-switch pulse generator went to all scintillator and counter inputs. During normal operation of the equipment, this generator gave one pulse each hour which was recorded in the same way as a normal event. This not only provided an hourly time marker, but also provided a calibration for the pulse-height scale, since the heights of the pulses from the generator were held constant to within ±0.5 percent throughout the experiment. Each calibration pulse also provided a check of the operation of the delay multivibrator circuits.

Approximately once a week an energy calibration of the pulses from the sodium iodide counter was made using the 4.44-Mev γ rays from a Po-Be source. The pulse-height distribution due to the γ rays was recorded and measured in the same way as in the cyclotron arrangement. The results of these calibrations showed that the gain of the photomultiplier remained constant within ±1 percent during the course of the experiment.

The recorded oscilloscope traces were projected onto a horizontal screen on a table-top with an enlargement such that the images were five times the size of the original trace on the face of the cathode-ray tube. Full scale deflection corresponded to about 20 cm on the screen, and the most probable pulse height to about 7 cm. The height of each scintillation counter pulse was measured with an error of ±0.01 cm and recorded, along with the information provided by the counter tray pulses. The artificial calibration pulses were also measured along with the genuine events. Each measurement of an event was converted to an energy loss in Mev by dividing by the average size of the calibration pulses at the time of the event and multiplying by the Mev equivalent of a calibration pulse as determined from the Po-Be calibrations.

RESULTS

The relation between the probable energy loss of a singly charged particle, ϵ_{prob}, in a thickness of t cm of material is expected to be given by the equation

$$\epsilon_{\text{prob}}=\frac{2\pi ne^4t}{mv^2}\left[\ln\frac{2mv^2(2\pi ne^4t/mv^2)}{I^2(1-\beta^2)}-\beta^2+j-\delta\right],$$

where n is the electron density, e is the electronic charge, m is the mass of the electron, I is the average ionization potential of the material, and v is the velocity of the charged particle. The term j is a small correction term calculated from straggling theory[14] which depends upon the absorber thickness and the particle velocity and which approaches 0.37 when

$$2\pi ne^4t/mv^2\ll 2mv^2/(1-\beta^2).$$

TABLE I. Calculated and observed counting rates.

Type of event	Calculated rate/hour	Observed rate/hour
E	0.518	0.586±0.041
EF	0.645	0.640±0.043
EFG	0.925	1.093±0.056
EFGH	2.31	2.12 ±0.077

[34] B. Rossi, Revs. Modern Phys. **20**, 538 (1948).

δ represents the density effect correction, which arises because the polarization of the medium reduces the energy loss. Numerical values of δ have been calculated by Sternheimer[10] for sodium iodide, which is the material used in this experiment. If $x=\log_{10}[\beta/(1-\beta^2)^{\frac{1}{2}}]$, then δ is given approximately by

$$\delta=0 \qquad x<0.09;$$
$$\delta=4.606x-5.77+0.278(3-x)^{2.77}, \quad 0.09<x<3.00;$$
$$\delta=4.606x-5.77, \qquad x>3.00.$$

Following Sternheimer, the mean ionization potential I was taken from an interpolation of the values given by Bakker and Segrè[35] to be 392 ev. In the cyclotron arrangement the correction for the lengthening of the average path length of the mesons in the sodium iodide due to scattering ranged from 0.66 percent at the lowest energy to 0.07 percent at the highest energy. In the cosmic-ray arrangement, the counter geometry (Fig. 3) was such that the average path length was increased by only 0.2 percent of the actual scintillator thickness of 1.51 cm. The theoretical values to be compared with the experimental results were all computed from the above relations.

While ionization loss is the only process which contributes significantly to energy loss by muons, this is not true for pions. In addition to ionization energy loss, pions may also lose energy by nuclear interaction. On the basis of emulsion, cloud chamber, and counter studies of pion interactions,[36] five percent of the pions are expected to undergo nuclear interactions and another five percent to be diffraction scattered in the 1.51 cm of sodium iodide. Less than five percent of the nuclear interactions will be counted, since a pion with $E>35$ Mev or a proton with $E>70$ Mev within 30° of the beam direction is required to actuate the coincidence counters (B and C in Fig. 1). The lengthening of the average path length in the sodium iodide due to the diffraction scattering of pions is less than 0.1 percent. For the one percent accuracy required in this experiment the effects of nuclear interactions and diffraction scattering can be neglected. Pions and muons also differ in spin, pions having spin zero and muons probably having spin $\frac{1}{2}$. However, the effect of the magnetic moment probably associated with the muon is completely negligible over the range of energies used in this work.[37] Therefore, since the energy loss equation for singly charged particles heavier than the electron involves only the velocity of the particle, pions and muons are expected to give equally valid results on the energy loss in sodium iodide.

Before stating the results, it is important to explain

[35] C. J. Bakker and E. Segrè, Phys. Rev. **81**, 489 (1951).

[36] Bernardini, Booth, and Lederman, Phys. Rev. **83**, 1075 (1951); R. L. Martin, Phys. Rev. **87**, 1052 (1952); J. O. Kessler and L. M. Lederman, Phys. Rev. **94**, 689 (1954).

[37] H. J. Bhabha, Proc. Roy. Soc. (London) **A164**, 257 (1938); H. S. W. Massey and H. C. Corben, Proc. Cambridge Phil. Soc. **35**, 463 (1939).

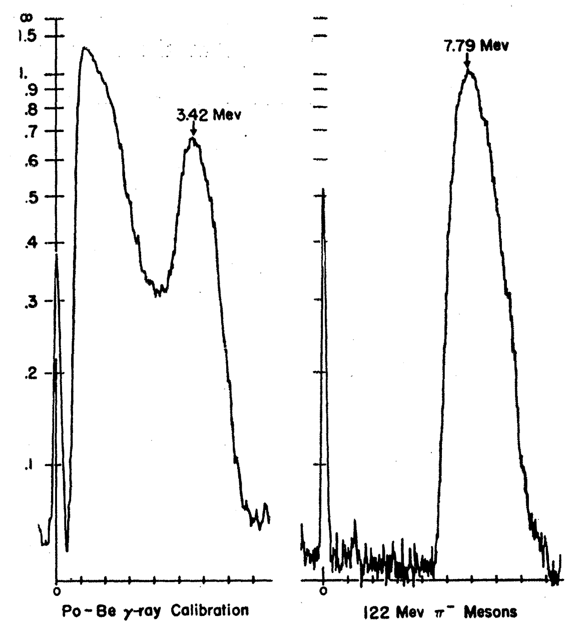

FIG. 6. Sample microphotometer tracings of the photographic density distributions due to (left) Po-Be γ-rays and (right) 122-Mev π^- mesons in sodium iodide.

how the most probable energy losses were estimated from the data. In the case of cyclotron-produced mesons, use was made of microphotometer tracings of the density distributions. Figure 6 shows typical density distributions for 4.44-Mev γ rays and for 122-Mev pions. Even though each distribution represents about 1.6×10^4 mesons, there are still small fluctuations in the curve. The position of the highest point of such a distribution curve clearly is not the best estimate of the position of the distribution. One can do better by also making use of the steeply rising and falling regions of the distribution curve.

In the procedure adopted for the cyclotron data, the most probable pulse height was determined with the help of a standard curve which was an average of a number of the observed distributions. The peak of this curve was marked. The standard curve was then superimposed on each pulse-height distribution to give the best fit possible by visual observation. The position of the peak of the standard curve was taken as the location of the most probable pulse height. By making use of the γ-ray calibration and assuming that the pulse height was proportional to the energy lost in the crystal, the distance from the zero reference line to the most probable pulse height could be used to calculate the most probable energy loss in the scintillator. The accuracy of a probable pulse-height measurement made in this way of an individual distribution was ±1.3 percent as determined from the consistency of all the results from a single energy. In using the above method there is the possibility of introducing a systematic error if the peak of the standard curve is in error, but this is less than one percent as judged from the consistency between results at the same meson energies from different runs.

A different procedure is required for finding the most probable energy losses from the cosmic-ray data, since each event is measured separately and the number of events is smaller than in the case of cyclotron-produced mesons. Over the range of energies covered by the cosmic-ray apparatus the energy loss distribution is expected to approach an exact Landau distribution.[13] The average pulse height is obviously an inaccurate estimate of the position of this distribution because of the long tail which causes the average of N measurements to be only slightly more accurate than a single measurement. A much more efficient estimate can be found by taking a modified median or quantile of the distribution. That is, since according to theory the most probable energy loss divides the area of a Landau distribution such that 28 percent of the area lies below and 72 percent above the probable loss, an estimate can be obtained from the data by finding the loss which divides the area under the experimental distribution curve in the ratio of 28 to 72. For the Landau distribution, the error in the probable loss obtained from this quantile method is about 1.5 times the error of the best possible estimate theoretically attainable.

A further difficulty arose when it was found that the actual energy loss distributions did not exactly fit the expected Landau distributions. The experimental

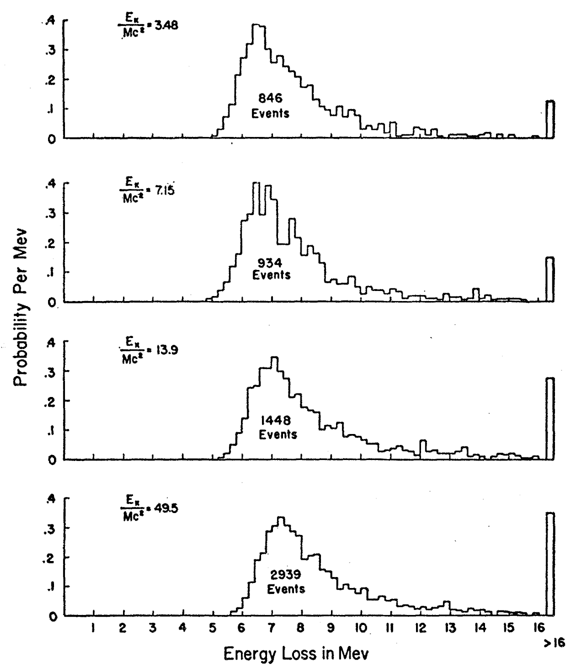

FIG. 7. Histograms of the cosmic-ray muon energy losses. The distributions from top to bottom represent E, EF, EFG, and EFGH events. The effective value of the muon kinetic energy in units of its rest energy (E_k/Mc^2) is given for each distribution. The shift of the histograms to larger losses for the higher energies can be seen clearly.

energy loss distributions are shown in Fig. 7. With increasing meson energy, more large energy losses were found than were expected. This was attributed to knock-on electrons produced in the scintillators and counter walls in the path of the mesons directly above the sodium iodide crystal. The energy distribution of knock-ons striking the sodium iodide crystal was calculated for several muon energies. The theoretical cross section for the production of knock-on electrons by muons (spin $\frac{1}{2}$) was used.[37] Since knock-on electrons produced with energies less than 1 Mev have very little chance of striking the sodium iodide crystal because of their large angle of emission and large multiple scattering, only electrons produced with energies greater than 1 Mev were considered, which made it possible to assume a constant rate of ionization energy loss in the calculation. These distributions were then folded into the Landau distributions. The resulting distributions had tails which agreed much better with the data, the number of large energy losses being more frequent at higher muon energies. The positions of the peaks of the original Landau distributions were not shifted by more than 0.5 percent, but quantile estimates of the probable losses would be shifted to larger values, the shift being dependent upon muon energy.

It was also observed that the peaks of the observed distributions were significantly broader than for those of pure Landau distributions. The theory of Blunck and Leisegang[15] predicts some broadening due to resonance losses in the neighborhood of the ionization potentials which for the sodium iodide crystal means that an additional fluctuation obeying a Gaussian distribution with a standard deviation of 0.10 Mev must be combined with the Landau distribution. Good agreement with the observed distributions requires assuming a standard deviation of 0.4 Mev. It is then most likely that the additional fluctuations arise from small nonuniformities in the light collection efficiency, statistical fluctuations in the number of photoelectrons and variations in gain of the photomultiplier.

Theoretical distribution curves, which included the effects of knock-on electrons and the Gaussian ($\sigma=0.4$ Mev) errors, were calculated. It appeared from these curves that a further improvement in estimating the probable energy loss could be obtained by cutting off the tail of the energy loss distribution. The cut-off was arbitrarily taken to be the position on a pure Landau distribution which divides the area so that 20 percent is in the cut-off tail. This cut-off was found to be 2.2 Mev above the probable energy loss. 35 percent of the remaining 80 percent of the area was found to lie below the probable loss and 65 percent above. The probable loss of an experimental distribution was obtained by starting with a crude estimate of the probable loss and placing the cut-off 2.2 Mev higher. An improved estimate of the probable loss was obtained from the quantile which divided the remaining area

TABLE II. Summary of results for the probable energy loss in 1.51 cm NaI. E_k is the average kinetic energy of the mesons.

Group	E_k/Mc^2	E_k (Mev)	Type	Energy spread (Mev)	Probable energy loss (Mev)	Relative error	Absolute error	Sternheimer's theory	Probability of least squares fit
122-Mev π^-	0.436	61	π	5	10.32	±0.08	±0.13	10.28	
Cyclotron	0.607	85	π	4	8.66	±0.06	±0.11	8.76	0.19
beam	0.843	118	π	3	7.77	±0.04	±0.09	7.83	
227-Mev π^-	0.857	120	π	7	7.84	±0.05	±0.09	7.82	
Cyclotron	1.164	163	π	6	7.19	±0.04	±0.08	7.23	0.55
beam	1.588	222	π	4	6.81	±0.03	±0.08	6.85	
	2.29	245	μ	5	6.50	±0.06	±0.09	6.59	
Cosmic	3.48	368	μ		6.48	±0.06	±0.09	6.53	
rays	7.15	755	μ		6.51	±0.06	±0.09	6.55	0.064
	13.9	1470	μ		6.78	±0.05	±0.08	6.72	
	49.5	5230	μ		7.13	±0.04	±0.08	7.02	

in the ratio of 35 to 65. The cut-off was then placed 2.2 Mev above the new probable loss estimate, the process of successive approximations going on until no further improvement was obtained. The modified quantile estimates obtained in this manner from the theoretical distribution curves were found to agree within 0.5 percent with the most probable losses given by the original Landau distributions. This method of arriving at the value of the probable loss has the distinct advantage of being unaffected by the presence of knock-on electrons or small Gaussian fluctuations in the distribution.

Table II gives the results from both the cyclotron and cosmic-ray data for the most probable energy loss at various energies. The cyclotron meson energies given are the average energies at the center of the sodium iodide (A in Fig. 1). Each cosmic-ray meson energy is that energy which would give the same probable loss as that expected from the actual spectrum of energies. This energy is very closely equal to the median of the respective energy band. The last row in the cosmic-ray group represents all muons with an energy greater than 2 Bev.

The relative errors of the probable energy losses are for use in making comparisons within one of the three groups into which the data has been divided. They refer to the errors in estimating the peaks of the pulse-height distributions, but do not include errors in the absolute energy calibration. The relative errors of the cyclotron results are estimated from the internal consistency of the data. The relative errors of the cosmic-ray data are computed assuming that the fractions of the data lying on either side of the true probable energy loss obey a binomial probability distribution. The error in the calibration needed to make absolute comparisons with the theory is estimated to be ±1.0 percent, to which ±0.6 percent is contributed by the uncertainty in the calibration γ-ray energy, ±0.5 percent by the error in locating the position of the distribution due to the γ rays, and ±0.3 percent by the uncertainty in the thickness of the sodium iodide crystal. An error in the calibration will affect all the losses in a group by the same factor, and is included in the estimate of absolute error, which should be used in comparing any single point with the theory.

The two master coincidences, $AB_1C_1D=M_1$ and $AB_2C_2D=M_2$ (see Fig. 3) allowed the data to be separated into two groups which passed through different regions of the sodium iodide crystal. The M_1 mesons passed through a region which was one inch closer to the photomultiplier than the M_2 group. The M_1 pulse-height distributions were located at 6.7 percent higher pulse heights than those of M_2. In the pulse-height histograms shown in Fig. 7, the M_1 distributions have been shifted down by this amount to coincide with the M_2 distributions. Except for this shift, the two sets of data agreed and showed the same relativistic rise within statistical errors. Use was made of this difference in light collection efficiency between the M_1 and M_2 groups in making comparisons with the calibration γ-ray distributions, which involve the whole volume of the crystal.

The results are also shown plotted in Fig. 8 along with a curve calculated from the theory of Sternheimer. For energies below the minimum of ionization, the

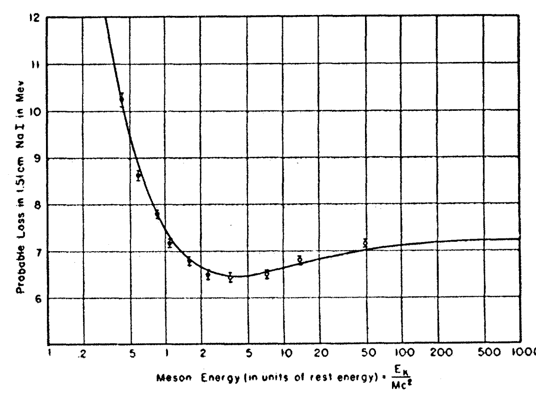

FIG. 8. The probable energy loss of mesons in 1.51 cm of sodium iodide is shown as a function of meson energy in units of its rest energy. The curve is calculated from the theory of Sternheimer. The solid circles represent cyclotron data using pions, except for the highest point at $E_k/Mc^2=2.3$, using muons. The open circles show the cosmic-ray muon results.

agreement between theory and experiment is perfectly satisfactory. Beyond the minimum the experimental data shows an indication of a somewhat more rapid rise, as can be seen in Fig. 8, where the errors indicated are the absolute errors of Table II. It is possible to test the agreement between theory and experiment in another way by making use of the relative errors, which are appreciably smaller than the absolute errors. The results from any one of the three groups in Table II can be normalized to the theoretical curve by a weighted least-squares fit. The exactness of this fit can then be tested by well known statistical methods. In the last column of Table II the probability due to statistical fluctuations of deviations as large or larger than those found is given for each group. The groups of data from the cyclotron give reasonably high probabilities (0.19 and 0.55), showing that the relative losses from theory and from experiment agree. The low probability of 0.064 for the points from the cosmic-ray data at energies in the region of the relativistic rise indicates a discrepancy between theory and experiment which may be significant.

To be more certain that the relativistic rise found in sodium iodide was not due to any instrumental errors, a preliminary check was made of part of the data from the liquid scintillator (terphenyl plus diphenylhexatriene in phenylcyclohexane) which was taken concurrently with the sodium iodide data. The probable pulse heights from all four range intervals agree and show that the relativistic increase could not exceed 2 percent, in contrast to the 10.9±1.0 percent increase found in sodium iodide. This result for phenylcyclohexane agrees with previous results on similar organic materials[22–24] and with theory.[10]

CONCLUSIONS

The results of this work demonstrate the possibility of exact determinations of the ionization energy loss of high-energy charged particles by the use of scintillation counters. Excellent agreement is found between theory and experiment for the probable energy loss of pions and muons at energies below the minimum of ionization where the density effect is not important. The experimental data for muons with energies beyond the minimum of ionization give conclusive evidence for the existence of the density effect in sodium iodide. In addition, the experimental results show a definite relativistic increase in the probable energy loss, the increase being 10.9±1.0 percent in going from the minimum to an energy of $50Mc^2$ ($M=207m_e$).[38] This compares with the prediction of 8.2 percent rise at $50Mc^2$ from the calculations of Sternheimer. The theory predicts a total rise in going from the minimum to the plateau at infinite energy of 11.4 percent, which is perfectly consistent with the results obtained here (10.9±1.0 percent) if it is assumed that the energy loss reaches the plateau more rapidly than expected.

It is a pleasure to acknowledge the advice and encouragement given by Professor Marcel Schein throughout the course of this work. The author also wishes to thank Professor H. L. Anderson and Mr. L. Kornblith for the use of the cyclotron and Dr. O. Joensuu of the Geology Department for the use of the microphotometer. Thanks are also due to members of the Cosmic Ray Group who have assisted in various phases of the work.

[38] Smith, Birnbaum, and Barkas, Phys. Rev. **91**, 765 (1953).