

PHOTOGRAPHIC EMULSIONS¹

By Y. GOLDSCHMIDT-CLERMONT

Laboratory Group, European Council for Nuclear Research, Zürich, Switzerland

INTRODUCTION

Because of its versatility, the detection of nuclear particles by photographic emulsions occupies an almost unique position among the various methods available for experimentation in nuclear physics. As in the cloud chamber, nuclear particles responsible for the tracks can be identified in a wide range of situations and their direction in space is readily determined. In addition, the photographic plate technique has a certain simplicity and, in its less refined forms, is inexpensive. The plates can be simply used as particle recorders, when it is sufficient to count the number of identical tracks occurring over a certain area. They are then in strong competition with more automatic detectors such as counters, but have the very definite advantage of easy discrimination, even, for instance, for detecting nuclear particles in a high background of electrons and γ -rays. This property makes them a first choice for exploratory experiments with particle accelerators of all energies. A number of measurements can be performed on the tracks themselves, and accordingly, provide the means of making the plate an increasingly refined instrument, adapted for instance to the detailed analysis of all tracks belonging to even extremely complicated nuclear events. In addition, combinations can be made of plates with other techniques, such as absorbers of various materials or deflection in a magnetic field, thus extending even further their field of usefulness. Other properties of plates may enhance their choice for experiments. Among these properties there are: small size and weight, ability to integrate events occurring over a long period of time, and relatively high density. The last property has the special advantage of quickly bringing to rest charged particles traversing them, enabling one sometimes to observe decay at rest of unstable particles (μ , π , τ , κ mesons), etc. Disintegrations of the nuclei of the emulsion itself are often observed, but the interpretation of these experiments is made difficult by the chemical complexity of the emulsion.

Very useful introductions to the basic techniques are found in the book of Rossi (1), in an article of Fowler & Perkins (2) and, with much information on procedures and applications, in a review article by Rotblat (3). Another review giving a broad picture is that of Beiser (4). An interesting historical account is found in a recent article by Vigneron (5), while early stages of the technique were reviewed by Shapiro (6) and, in more restricted fields, by Demers (7). The reader will certainly find a reward in looking at the beautiful microphotographs of the book by Powell & Occhialini (8). Much information

¹ The survey of literature pertaining to this review was concluded in February, 1953.

on the use of emulsions for radioactive measurements, low-energy reactions, and applications to various fields, is found in a book by Yagoda (9). Many applications to the field of meson physics are reviewed by Powell (10).

The scope of the present chapter is limited. Only the measurements to be performed on the tracks themselves are examined. In addition it has been thought useful to include a discussion of processing techniques which are the basis of any successful experiment. The emphasis is mostly centered on the major progress of recent years, the use of more sensitive emulsions in great thicknesses, extending the application of the method to the high-energy region. But the author does not feel competent to do justice to what are perhaps the most fundamental aspects of the technique, those connected with the photographic process itself. No account is given, for instance, of the problems of emulsion manufacture for which the makers of emulsion have given the physicists indefatigable help. Yet the importance of research in the difficult fields of the mechanism of latent image formation, the factors affecting sensitivity, and the interpretation of development and fixing processes cannot be overemphasized. In their study may lie the key to radical improvement of the technique. The reader will probably find the best introduction to these questions in the very interesting review of Waller (11). Some of the material of this chapter of necessity parallels that of the reviews already quoted, although duplication is avoided when it is felt that the reader can be referred to them for a more complete treatment than space limitations would here allow. No mention is made, for instance, of the simplified processing methods which can be used in the lower energy region with thinner plates of less sensitivity (3, 4). The numerous precautions necessary to achieve good microscopic observations and measurements are not discussed (3, 12). Nor is there discussion of specific uses of the plates for the detection of neutral particles (3, 4), for applications to nuclear physics (3, 4, 8, 9), cosmic rays (13, 14), geology (11), etc.

The methods available today for the use of emulsions result from painstaking efforts of many workers. Significant improvements are often communicated from one laboratory to the others by way of visits or friendly letters. As a result, it is difficult to give always proper credit to authors by bibliographic references, which should be interpreted, when given in the present text, as guidance only to more detailed treatment.

PROPERTIES OF EMULSIONS

Emulsions sensitive to tracks of minimum ionization are manufactured in the United States by Eastman Kodak [type NTB 3 (15)], in England by Kodak [type NT 4 (16)], and by Ilford [type G 5 (17)]. Some authors have made their own emulsions (18, 19). As an illustration, Table I lists some of the properties of the Ilford G 5 emulsions. The reader will find data on a number of emulsions of different sensitivities, some of them loaded with specific elements, in the data sheets published by the manufacturers (15, 16, 17, 20) and in the review articles already quoted. It is of interest to note

that Ilford produces several types of "diluted G 5" emulsions, sensitive to minimum ionization, where the gelatin content has been increased, thus substantially augmenting the proportion of light elements (20, 21). Manufacturers are most helpful in supplying physicists with plates of almost all sizes and shapes upon request, although they have established convenient standard sizes. The thickness of the emulsion commonly used has much increased in recent years; Ilford reports (20) that 400, 600, 1000 and 1200 μ emulsion layers are produced regularly and that 2000 μ has occasionally been made. Also available are emulsions without glass backing, sometimes called "pellicules" or "stripped emulsions," which are very useful, as they can be stacked into a large sensitive volume. The relative position of the layers is usually ascertained by exposures to thin pencils of x-rays to facilitate the observation of the successive segments of tracks in each pellicule after processing. Ilford (22) also supplies emulsion in gel form and treated glass for special applications when it is essential that the experimenter pour his own plates to avoid any preliminary exposure.

TABLE I
DATA ON ILFORD G 5 EMULSIONS*

Composition †			
Density	3.907 gm./cm. ³	Silver	1.85 gm./cm. ³
Atoms/cm. ³	8.12×10^{22}	Bromine	1.36
Mean A	28.98	Iodine	0.024
Mean Z	13.17	Carbon	0.27
Mean Z ²	456	Hydrogen	0.056
Radiation length	2.93 cm.	Oxygen	0.27
		Sulfur	0.010
		Nitrogen	0.067

* In equilibrium with an atmosphere of 50 per cent relative humidity.

† From Ilford Research Laboratories (17).

The effects of pressure, temperature, humidity, etc. on emulsion properties and on latent image stability are described in detail by Beiser (4) and can be examined only briefly here.

Temperature.—The emulsion melting point is somewhere above 45°C. and the plates should, of course, not be exposed to excessive heat. The influence of temperature on sensitivity in Ilford G 5 emulsions was studied by Lord (23) who found the maximum sensitivity around 0°C. with slightly decreased sensitivity at higher and lower temperatures. The emulsion is still sensitive to minimum ionization at -60°C. It seems to have zero sensitivity at -200°C. The temperature effect may be higher in less sensitive emulsions (24).

Pressure.—Although early types of plates were quite sensitive to surface pressure, this is no longer the case. Upon request, some manufacturers have

added a thin coating of transparent gelatin to the surface of their plates as a protection against scratches etc. Plates can be exposed at low pressure and in vacuum, but the loss of humidity which then occurs can make the emulsion leave the glass, unless ordered from Ilford "with extra plasticizer" [some change in composition (20)] or dipped in a solution of glycerine [recommended by Eastman Kodak for their emulsions (15)]. The author exposed Ilford G 5 emulsions in nitrogen of 50 atmosphere pressure and more, without apparent effect.

Humidity.—The hygroscopic properties of gelatin make the density of emulsion depend on atmospheric humidity. The manufacturers supply data on the influence of humidity on their plates. The corresponding change in composition in emulsion thickness, in stopping power etc., must be taken into account for precision measurements. Humidity has also a slight influence on sensitivity.

Contact.—Contact with metal like copper, aluminum, iron, etc., fogs, or even decomposes the emulsion. A similar effect is observed when exposing the emulsion in hydrogen, in which case complete blackening of the plates occurs quite rapidly unless the plate is cooled to a sufficiently low temperature (25). It seems very difficult to find a thin coating which can be applied to the surface of the emulsions and through which hydrogen can not diffuse.

Fading.—The mechanism of the fading of the latent image with time and the influence of physical and chemical factors on this phenomenon were the subject of much research (4). Although fading is often negligible in minimum ionization emulsions, it is increased by higher temperature and humidity, so that such conditions should be avoided as much as possible during exposure and before development.

Eradication.—Accelerated fading is possibly the best available method for eradication of the latent image before exposure, but it is sometimes difficult to achieve without reducing the sensitivity of the plate. Albouy & Farragi (26) report that latent images in Ilford G 5 are destroyed without effect on sensitivity by heating for a few hours in a humid atmosphere at temperatures ranging from 60 to 90°C. The plates must, of course, be kept strictly horizontal. Other interesting results are those of Goldstein & Sherman (27) on the use of Herschel effect.

Shrinkage.—After processing, the thickness of the emulsion is changed and usually reduced by a factor of the order 2 or 3. This shrinkage sharply depends on the concentration of the glycerine bath used at the end of the processing (see below) and on the atmospheric humidity during observation (12). Various methods are available to measure this effect (28, 29).

Proper processing makes the images of minimum ionizing particle appear, in a sensitive emulsion, as rows of developed grains with densities of 20 to 30, or even more, grains per 100 μ . The visibility of these tracks depends on the ratio of the grain density of the tracks to the surrounding background of slow electrons and randomly developed grains (fog). Interesting studies of track recognition in varying conditions of grain and fog densities are given

by Coates (30), Berriman (31), and Beiser (32). More ionizing particles produce denser tracks: first with increased grain density, and then with the appearance of continuous strings of increasing width. The tracks are usually not straight lines, but show multiple scattering. Electrons can be ejected from atoms situated near the path of sufficiently high-energy or highly charged incident particles, and appear as individual δ -rays. Slow particles often are brought to rest and end their range in the emulsion. Multiply charged particles capture electrons toward the end of their range, thus decreasing in effective charge and giving a characteristic tapered shape to their tracks. Quantitative measurement and theory of these phenomena are at the basis of the photographic emulsion technique, as they permit the determination over certain ranges, of the charge, mass and energy of the incident particles.

PROCESSING

For all experiments where measurements need to be performed on the tracks and where a low background (γ -rays, electrons etc.) can be maintained, there is a premium in having long tracks, so that emulsions of great thickness must be used. The central problem of processing thick emulsions is to achieve an even development throughout the thickness and over the whole area of the plate, in spite of the fact that developers and other chemicals only penetrate by a relatively slow diffusion process from the surface. Other stringent requirements are a high contrast, i.e., the tracks easily recognizable from a background of random grains (fog as low as possible) with no coloration of the gelatin; and absence of distortion, so that to the highest degree of precision the developed grains reproduce the original paths of the charged particles. Distortion can occur in all stages of the processing, if proper precautions are not taken. It is essential, for instance, that the plates remain horizontal, that at no time strong gradients of concentration, or temperature exist in the emulsion or its vicinity, and that any osmotic pressure effects be reduced to a minimum. It is well known that the swelling of the gelatin is a function of its pH and, it may well be that the ideal development should be made in successive baths of constant pH. It is convenient to divide the processing technique in two steps: (a) the developing stage, where the plate is usually soaked in distilled water to allow some swelling, and then through suitable chemical action the sensitized silver bromide grains are reduced to metallic silver, after which the action of the developer is checked by a neutralizing agent; and (b) the after-developing stages including the fixing where the undeveloped opaque silver bromide is dissolved, the washing and the drying.

Development.—The simplest method to achieve even development of thick emulsions is perhaps to choose a slow developer with a long induction period. It penetrates throughout the thickness in a time appreciably less than the developing time itself. Standard x-ray film developer, like the D 19, fulfills this condition when suitably diluted for thicknesses up to 100

or 200 μ . Recently Summerfield (33) reported an extension of this simple method to thicknesses of up to 400 μ by using a relatively slow developer, and further slowing down its effects by reducing the temperature to 4°C. With this technique, developing, stopping and fixing are all done at the same low temperature with the additional advantage of keeping the gelatin in a relatively firm condition and thereby reducing the possibility of distortions.

The most widely used method to-day is the Temperature Development technique (TD) of Dilworth, Occhialini & Payne (34), further studied by numerous workers (35 to 42). The TD technique is based on the difference in temperature coefficients between diffusion and developing processes. A developer is chosen whose chemical effects are more reduced at low temperature than is its diffusion speed. The development begins by a bath in distilled water which produces a preliminary swelling of the gelatin and facilitates the subsequent diffusion of chemicals. The duration of this preliminary bath can be the same as that of the cold stage discussed below. The bath is slowly cooled. Then begins the "cold stage": The distilled water is replaced by a cold bath of the developer which is allowed to diffuse evenly throughout the thickness of the emulsion, while no appreciable development occurs. The plate is then taken out of the bath, the excess developer on its surface is gently wiped off, and during a "hot stage," the plate is slowly heated in a humid and sometimes inert (nitrogen) atmosphere up to a temperature where the chemical developing effects occur. The developer, imbedded in the gelatin, should then produce identical effects in all layers, in particular, the surface, not being in contact with fresh developer, does not receive preferential treatment. After a suitable time, the temperature is lowered to stop developing effects and the plate is immersed in a cold stop bath. This method has proved successful for thicknesses up to 1200 and even 2000 μ .

The requirements for a developer suitable for the temperature development technique are quite stringent and not easy to meet. Quick penetration through emulsion, moderate or high temperature coefficient, stability at high and at low temperature, negligible production of fog or stain, pH close to that of emulsion, no solvent effect for silver bromide, absence of oxidation by air are some of the conditions listed and examined by Dilworth, Occhialini & Vermaesen (35). Among the large number of known photographic developers there are certainly candidates for filling several of these requirements, but it is hard to find one that meets them all. Much research has already been devoted to this choice, and amidol (2:4 diaminophenol hydrochloride) seems to be the best available at present. It is used in a solution of proper preservative, restraining and buffering qualities, the composition of which may vary according to the thickness of emulsion processed. In particular, when thick emulsions are developed, the cold stage is very long, and the developer must have restraining properties not obtained with the usual addition of potassium bromide. More restraining effect can be obtained by making the solution slightly more acid. Two mild acidifying agents were

proposed for this purpose, boric acid (35) and sodium bisulphite (37) which also improve the buffering qualities of the developer solution. The composition of two of the more widely used developers is given in Table II. As they are rather rapidly oxidized by air, the developing solutions should be prepared immediately before use and only used once.

TABLE II
COMPOSITION OF TYPICAL AMIDOL DEVELOPERS

	Brussels*	Bristol†
Amidol	4.5 gm.	3.0 gm.
Sodium sulfite (anhydrous)	18 gm.	6.7 gm.
Potassium bromide (10 per cent solution)	8 cm. ³	
Boric acid	35 gm.	
Sodium bisulfite liquor (Specific gravity 1.34)		1.4 ml.
Distilled water	1000 ml.	930 ml.
pH	6.4	6.7

* From Dilworth, Occhialini & Vermaesen (35).

† From Dainton, Gattiker & Lock (37).

The temperature of the cold stage does not seem to be very critical, most authors use 4 or 5°C. or less. The time required varies with thickness of emulsion. Dainton, Gattiker & Lock (37) measured penetration time of various developers, including amidol, in nuclear emulsions. However, longer times are usually recommended and a good working rule is 20 to 25 min. for each 100 μ of emulsion thickness. The temperature of the hot stage is very critical, and a slight change can produce an appreciable difference in degree of development. It is useful to control the rates of temperature rise and drop at the beginning and at the end of the hot stage to insure accurately reproducible results. These rates should not be too fast, otherwise distortion or even reticulation of the gelatin may occur. It is also found that the temperature needed for the hot stage varies with emulsion thickness, and also slightly from batch to batch. These properties necessitate use of apparatus to control temperature accurately and to insure even temperature distribution over the whole area of the plate. Proper choice of hot stage temperature affords a convenient method of obtaining any required degree of underdevelopment for special purposes. The boric acid type of amidol developer seems to require somewhat higher hot stage temperatures than the bisulphite type. Indeed, the bisulphite developer used at 10 or 14°C. for a long enough time provides enough development to show minimum ionization tracks in Ilford G 5 plates, although the grain density is not as high as may be desired [Herz (39)]. Herz & Edgar (41) point out the advantages of using a lower temperature for the hot stage: when the wet emulsion is warmed, it loses elasticity and becomes dough-like and sticky, stresses

are released if present, and when the emulsion is later dried, it has a much greater tendency to shrink in a direction parallel to the glass base than one which has been kept at a lower temperature throughout. The boric acid amidol and the temperature development technique produce images of very good contrast and extremely low amount of distortion when applied with proper care, using a hot stage temperature of about 28°C. for 30 min. to 1 hr. Alternatively, excellent results are also obtained by using a much lower hot stage temperature, of 8°C. to 14°C. for several hours, with the advantage that absence of distortion may be more easily achieved.

Special apparatus is needed to supply the solutions at required temperatures and to perform the necessary temperature changes with accuracy and reproducibility. For infrequent development of relatively few plates; one of the simplest of these, is a hermetic container of suitable geometric design to insure even heat transmission and temperature distribution, which can be immersed (with plates and solution inside) in successive water baths of suitable temperature. For frequent developments of large numbers of plates, in strict conditions, more elaborate apparatus are needed, such as that described by Dilworth *et al.* (35). Essentially it consists of a jacketed container for plates and solutions with an automatic device providing a circulation of water of controlled temperature in the jacket.

The composition of the stop-bath is not very critical. Usually a slight change in pH, making the plate more acid, is sufficient to stop completely developing effects, and very dilute acetic acid or sodium bisulphite is used. The duration of the stop-bath is governed, as the cold stage, by the time required for the diffusion process to take place, and again an empirical rule of 20 to 25 minutes for each 100 μ of emulsion thickness can be recommended.

After development.—After the stop-bath, it is convenient to remove the thin silver layer sometimes deposited on the emulsion surface by rubbing gently with a soft tissue, and the plate is then washed in running water of low temperature to eliminate as far as possible all remains of used developer and stop-bath. The plate is then ready for fixing. The fixing is amongst the most critical phases of processing, as the large fraction of undeveloped silver bromide must be dissolved and evacuated. "In the development stage, the emulsion, held firm only on the side of the glass, has the support of a hard skeleton of silver halide, whereas during fixing, washing and drying it is reduced to a soft jelly (43)." The choice of the composition of the fixing bath has received much attention as it appears to have a major influence on the final transparency of the plate. The basis of all fixing baths to date is sodium thiosulphate (hypo), used at about 40 per cent concentration. Dainton, Gattiker & Lock (37) and other authors (42) recommend the addition of sodium bisulphite, making the solution more acid and therefore reducing the swelling and to some extent the staining of the gelatin. This practice is not followed by Bonetti, Dilworth & Occhialini (43) who prefer not to decrease too much the pH of the hypo solution to avoid the risk of decomposition. They control the amount of swelling by observing the thickness of a control

test plate with each processing, and by the progressive addition of sodium sulphate up to about 10 per cent concentration. They use a bath of very dilute acetic acid to remove staining after the fixing is completed. A difficult question is that of agitation and solution renewal during fixing, as any excess of motion and any brutal change of concentrations may cause distortion, while complete absence of motion and local or total exhaustion of the solution increase the fixing time and the staining. Nitrogen bubbling can provide some agitation (36). An apparatus producing an even and gentle laminar flow of hypo above the plates is described by Bonetti, Dilworth & Occhialini (43).

It is essential to end the fixing by progressive dilution of the hypo at a very slow rate. Rapid changes of concentration at this stage are known to produce severe distortion and may even cause the emulsion to leave the glass. The dilution may start when the plate first looks transparent and, as dilution proceeds, fixing is progressively completed. It can sometimes be followed by a bath of dilute acetic acid which may help to reduce the staining. The plate can then be washed and is ready to dry. It is recommended, before drying the plates, to give them a bath in a solution of glycerine at a concentration

TABLE III
EXAMPLE OF TEMPERATURE DEVELOPMENT SCHEDULE FOR 600 MICRONS PLATE

Operation	Bath	Temperature	Time
Development			
Preliminary soaking	distilled water	cooling down to 5°C.	120 min.
Cold stage	boric acid amidol*	5°C.	120 min.
Warm stage			
slow heating	dry†	5°C. to 28°C.	5 min.
development	dry	28°C	60 min.
slow cooling	dry	28°C. to 5°C.	5 min.
Stop bath	acetic acid 0.2 per cent	5°C. to 14°C.	120 min.
Silver deposit cleaning‡			
After development			
Washing	running water	14°C.	120 min.
Fixing	hypo 40 per cent§	14°C. cooling down to 5°C.	until clear
Slow dilution	water§	5°C.	100 hr.
Glycerine bath	glycerine 2 per cent	5°C. to ambient	120 min.
Drying		20°C.	7 days

* See Table I.

† Wipe the plate surface with a soft tissue.

‡ Remove the silver deposited on the surface with a soft tissue.

§ Add sodium sulphate in increasing concentration up to 10 per cent if swelling is excessive.

of a few per cent. This will prevent the emulsion from becoming too dry (when the humidity of the laboratory atmosphere is low) and from breaking its glass backing. It is of interest to note that the degree of shrinkage of the emulsion strongly depends on the concentration of glycerine, so that it can be controlled adequately (44, 45).

The drying can be made by immersion in a succession of baths containing glycerine and alcohol in increasing concentration. If carried up to very high concentrations of alcohol, it may give unwanted hardness to the gelatin (43). The other method of drying relies on evaporation, its main difficulty is to achieve even evaporation over the whole area of the plate. This can be obtained by controlled slow air circulation, but special precautions are needed for the edges of the plate which have the tendency to dry first. The plates can be surrounded by a guard ring of dummy plates (43) or very large plates can be used of which an area near the edges is discarded (37). It is possible to use gentle infrared heating to help the process of drying (43). As an illustration, a typical development schedule for Ilford G 5 plates of 600 μ is given by Table III.

Several essential techniques cannot be discussed here in detail: developers giving a controlled degree of underdevelopment (46); developers selectively developing track images in a background of γ -rays (47, 48); a method for clearing overstained plates (49) (a result which can also sometimes be obtained by a new fixing after the plate is dried); a procedure for mounting stripped emulsions on glass, before processing (40, 50); and the quantitative measurements of distortion (51, 52).

OBSERVATION

Like all other experiments, those that can be performed with nuclear emulsions encounter some of their most severe limitations when they meet the limitations of the human being using them. He can only handle a limited amount of information over a given time and experiments must usually be done over a few months or years. Scarce are the experiments with plates where more than a few thousand tracks were examined. Observation of the plates under the microscope is, of course, where human limitations appear. Scanning of an emulsion to locate events requires long hours of patient observation and is usually entrusted to professional scanners, recruited from semi-skilled personnel and patiently taught to use the microscope, to recognize events and even to perform measurements. Scanners thus become extremely useful assistants and a group of well trained professional scanners may be the most valuable asset of many laboratories using the technique. Even with their help, physicists must spend hours at their microscope, looking at tiny details of the field of view, with head in a fixed position in front of the eye-pieces and steady hands delicately moving the mechanical controls of the instrument. This is often found too taxing and may be the main reason for the technique not having a still more extensive use.

Any improvement in microscope design is worth watching. Much re-

search effort has already been devoted to improvements of this instrument. Most of the microscopes available commercially are built for biology and medicine, with various degrees of mechanical perfection of the stage. Only the most perfected of these find their use for photographic emulsion technique and then, are not very well adapted except for the simplest operations, for instance, scanning and location of events. To perform measurements on the tracks the graduations of the stage are usually not precise enough and the motions are actuated by too coarse screws. It appears that, as for mechanical construction of microscopes, the domain of emulsion technique starts just about where the technique in medicine and biology stops. Microscope manufacturers have only just begun to explore this new domain. Some of them have supplied physicists with special accessories custom-built for a special need, such as a precision screw, allowing an accurate determination of distances (53). A remarkable pioneering design of microscope, specially built for nuclear emulsion technique, has been made by Cooke, Troughton & Simms, in England. Their model 4000 found, so to speak, its use mostly limited by price for a long time. It is still today one of the commercially available microscopes best adapted for emulsion work. Another special microscope is now being manufactured by Koristka, in Italy, based on Cosyns' principle (54), and especially suited for the measurement of multiple scattering. Other models were made or announced by Bausch & Lomb in the United States and Leitz in Germany. In the opinion of the author there is still room for much development of special microscopes for emulsion technique. This may be illustrated by reference to a number of convenient accessories that many authors have built themselves in laboratory workshops, such as tilting stages (55), or tilting microscope bodies (45), precision screws (56), recording devices (55, 57), goniometers of high accuracy (54), also stages with good rectilinear motion for measurement of scattering (54, 58). If some of these accessories, and perhaps others, could be perfected and made readily available to the users of emulsion technique, there is little doubt that they would soon find a relatively broad market.

Similar considerations apply to the optical system. The observation of thick emulsions requires objectives with medium or high magnification, flat field, large numerical aperture, and large working distance (distance between the object plane and the front lens). Several objectives meeting these requirements were successfully developed in recent years by Cooke ($\times 45$, n.a. 0.95, w.d. 1.5 mm.), Leitz ($\times 53$, n.a. 0.95, w.d. 1 mm.), Koristka ($\times 55$, n.a. 0.95, w.d. 1.35 mm.) and others. These are most useful for observation and measurements of selected events. For systematic scanning the need was long felt for an objective with low magnification and high numerical aperture using oil immersion. This need was recently filled by Leitz ($\times 22$, n.a. 0.65, w.d. 2.3 mm.) and Koristka ($\times 30$, n.a. 1.05, w.d. 3 mm.). These objectives solve the old difficulty of constantly changing from dry objective to oil immersion. In addition, the use of oil greatly improves visibility when, as is often the case, the surface of the emulsion is not perfectly smooth. New improvements

in optical systems are hoped for, such as a wider field of view at low power. It may be useful to remark that optical systems for nuclear emulsions can be designed without chromatic corrections, as the examination is easily made in monochromatic light.

Several attempts were made to make the microscope somewhat automatic. Successful instruments were built for limited purposes: to facilitate scanning (59, 60, 61), to record observation (57), or to make automatic measurements (62). One of the main difficulties in the task of making automatic microscopes may be to draw a line between the opposite requirements of versatility and mechanical simplicity. Another is to devise improvements that really relieve the task of the observer who must look at the tracks and guide the automaton. The most elaborate instrument recorded in the literature is perhaps that of Blau, Rudin & Lindenbaum (62). In spite of these very interesting efforts, few automatons were used for actual investigations in physics: an indication that the problem is difficult and that the field is still open for much progress.

The human eye has been replaced in measuring the density of tracks by a photometric device (described in Energy Loss and Measurements of Ionization). However, the eye has not been replaced in its role of locating events. Experiments have been performed in which the microscopic image is scanned with a television system thereby translating the field of view into a succession of electrical impulses (63, 64). This problem is made difficult by the high resolution needed and by the fact that the microscope itself is often used at the limit of the optical resolving power. Even if the scanning problem were satisfactorily solved, a computing mechanism is required to decide whether or not some of the grains among the many grains scattered over a field of view are aligned over the straight or slightly curved line of a track (63, 64), while even the most inexperienced eye will detect them at a glance. The same difficulty holds even more for the selection of a given type of complicated event. An electronic computer can probably be taught to recognize, say, a meson decay, but it will take much development and research before electro-mechanical scanning robots will contribute to advances in nuclear physics.

RANGE-ENERGY

The range of a charged particle brought to rest in the emulsion can be measured with high accuracy, and a considerable amount of theoretical and experimental work has been devoted to establish a precise range-energy relation. In principle, this relation can be calculated by integrating the Bethe (65) formula for energy loss, provided proper values of the average ionization potentials of the atoms are inserted. Improvements were made recently in the experimental knowledge of the potentials (66) and, in stopping power theory, in the calculation of the contribution of the K electrons (67, 68). A critical survey of their use for emulsion is made by Vigneron (69), who gives a semi-empirical treatment of the range-energy relation in excellent agreement with experiment. This calculation also agrees with earlier results,

based on the range-energy relation in air, and on a calculation of the ratio of stopping power between air and emulsion [Cüer (70), Webb (71), Wilkins (72)]. Extensive experiments have established the range-energy relation for protons and other singly charged nuclear particles. The early results of Lattes, Fowler & Cüer (73) were improved by Rotblat (74), Catala & Gibson (75), Nerenson & Reines (76), Cüer & Jung (77) and others (78, 79), and extended to higher energies by Bradner *et al.* (80). A range-energy curve for dry Ilford C 2 emulsion exposed in vacuum is given in Fig. 1. Plates in equilibrium with the atmosphere at average pressure and humidity exhibit ranges increased by 3 to 4 per cent (74). No appreciable change is found between different batches of emulsion of the same type, but the stopping powers of type Ilford E 1, Ilford G 5, and Kodak NT 2a are respectively 0.3 per cent lower, 1 per cent higher, and 1.8 per cent higher than that of Ilford C 2. Range-energy relations are given for Eastman NTA (81), for Eastman NTB (82), for diluted Ilford G 5 (83), and for Ilford C 2 plates soaked with water (4, 72). The experimental data for protons between 14 and 30 Mev are represented with an accuracy of the order of 2 per cent by the simple approximate formula (80)

$$E_{\text{Mev}} = 0.251R_{\mu}^{0.581}$$

which can probably be safely extrapolated to energies as high as 200 Mev, as checked indirectly by Gottstein & Mulvey (84).

From the proton range-energy curve $R=f(E)$, the range-energy relation for other singly charged particles of M proton masses is easily derived by the familiar relation $R/M=f(E/M)$. The somewhat different case of electrons was investigated by Ross & Zajac (85), Hertz (86), and Blum (87).

The range of particles of M proton masses and charge z can also be derived from the proton relation, after a correction to take into account the capture and loss of electrons which occur when the particle slows down to atomic electron velocities. From his experiments with Li^3 and Be^3 nuclei and by comparison with earlier values (72) on α particles, Barkas (88) gives the following relation for light nuclei: $z^2R/M=f(E/M)+0.12z^3$ valid for $v/c>1.04z/137$. Somewhat different results are derived in a review by Longchamp (89). For heavier nuclei, the reader is referred, for instance, to the paper of Perkins (90).

It can be concluded that a precise measurement of ranges yields an accurate determination of particle energies. This holds true for the mean range of a group of monokinetic identical particles. For an individual track, a slight uncertainty is attributable to straggling. Values of the straggling, both resulting from statistical fluctuations in the slowing down process and distances between grains with finite size, are found, for instance, for C 2 emulsions (74). Straggling amounts to 10 per cent for 1 Mev protons with a decrease to 2 per cent for 5 Mev protons. At higher energies, straggling can usually be neglected being no greater than the uncertainty of the range-energy relation itself.

Taper length.—The ionization along tracks of multiply charged particles

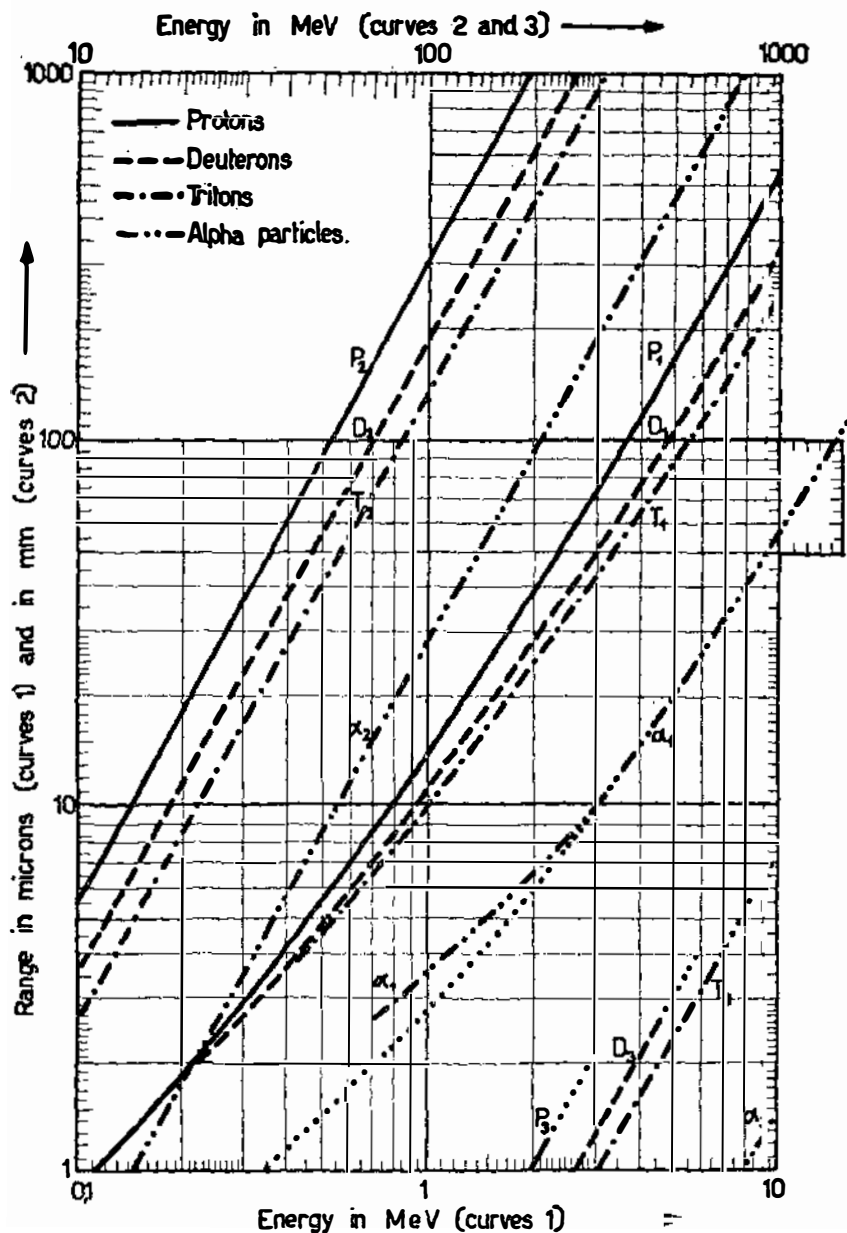


FIG. 1. Range energy relation for Ilford C 2 emulsion. From Vigneron (69).

show a characteristic taper at the end of their range corresponding to gradual loss of charge by electron capture (91). The taper length L can be expressed in function of the charge z by $L = az^\alpha$. The values for a and α result from an analysis of the ionization and electron capture phenomena. Perkins (90) used $a = 0.7$ and $\alpha = 2$, but experiments by Hoang (92) and supporting theoretical considerations of Cüer & Longchamp (93) indicate that α is of the order of unity.

ENERGY LOSS AND MEASUREMENTS OF IONIZATION

It is most likely that only a small fraction of the energy lost by a charged particle traversing the emulsion is used for the production of latent images that can be brought out during the development. The number of developed grains is a function not only of the rate of energy loss but also of the sensitivity of the emulsion and of the nature and degree of development. Grain counting and other techniques, discussed in some detail below, can provide experimental determinations of the rate of energy loss, which then can be examined in the light of theory to give information on the velocity and charge of the particle responsible for the observed track.

For slow particle velocities, the rate of energy loss is given by the well known Bethe formula, already mentioned in connection with range-energy calculations. The rate of energy loss is proportional to the square of the particle charge and a function of the particle velocity (apart from a negligible correction). The formula shows a decrease in energy loss as the particle velocity increases, with a minimum occurring at an energy of about twice the particle rest mass. After the minimum, the energy loss rises logarithmically with particle energy.

It is well known since the work of Fermi (94) that the rate of energy loss of high velocity particles is affected by the polarization of the medium. Negligible at low energies, this effect becomes considerable for relativistic velocities. Instead of a logarithmic rise, the rate of energy loss is found to saturate at high energies, at a value depending on the dielectric properties of the medium, but usually not very much above the minimum. Improvements were brought to the Fermi theory by Wick (95), Halpern & Hall (96), and Bohr (97), using more refined representations of the dielectric properties of the medium. Examining the applications of these theories to the grain formation in emulsions, Messel & Ritson (98) and Schönberg (99) pointed out that, since the energy is lost by fast particles in two distinct processes, ionization excitation and emission of Cerenkov radiation, the contribution of these processes to latent image formation along the particle track will depend on the probability that energy lost in these two forms is captured in the silver bromide crystals. As for the ionization process, electrons ejected with high velocity will appear as δ -rays with ranges much greater than the radius of the silver bromide crystals, in which they will spend only a small fraction of their energy. The energy lost in the form of such δ -rays will not contribute to latent image formation on the track. The quantity to be used for com-

parison with observed grain density is not the total rate of energy loss, but the energy lost in all collision processes involving electron energies less than some maximum energy value $E_{\max}$. This quantity is little affected by the exact value of $E_{\max}$ [evaluated to lie between 2 and 5 kev (98,100)]. Whereas the total increase predicted by theory in silver bromide amounts to about 30 per cent, the $E_{\max}$ limitation reduces the increase to about 15 per cent (see Fig. 2). These authors further demonstrated that the remaining increase was entirely attributable to increasing Cerenkov emission. This radiation is

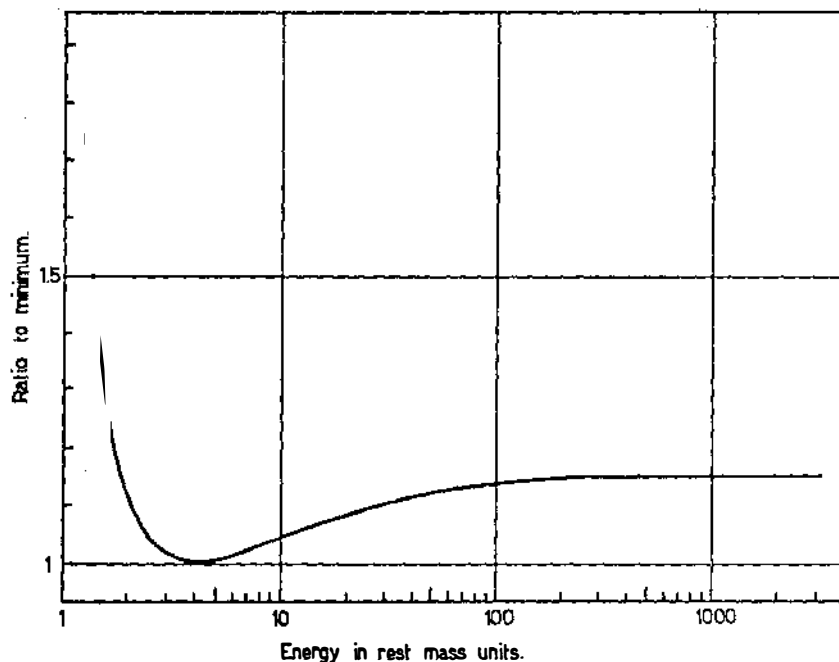


FIG. 2. Rate of energy loss in silver bromide in processes involving energy transfers of less than 5 kev., as a function of the particle energy. Calculated by Shapiro & Stiller (109) from results of Halpern & Hall (96) and Sternheimer (110).

emitted at frequencies in the neighborhood of resonance lines and is therefore strongly absorbed. But at least for the highest Cerenkov bands, it is very doubtful that the absorption coefficient would be high enough to stop the radiation from leaving the silver bromide crystals. Specific calculations on Cerenkov radiation capture in the crystals of silver bromide were made by Janssens & Huybrechts (100) following the theory of Schönberg, with the conclusion that only a small fraction was captured. Thus they were led to the conclusion that practically no increase in grain density was expected after the minimum.

This result seemed in agreement within experimental errors with early measurements (101, 102) but was not further confirmed experimentally.

Careful observations of the grain densities of fast electrons, mesons and protons established a definite increase of grain density between a minimum at about 2 rest masses and a plateau at high energies. Although the experimental errors are still appreciable, the results are not entirely consistent in two respects: the amount of increase and the value of the energy at which saturation is reached. Thus Pickup & Voyvodic (103) observing fast electrons, first measured a rise of about 10 per cent to a plateau starting at an energy of about 20 rest masses. These very preliminary results were later improved by observations of Morrish (104) on fast electrons, who detected a rise of about 5 per cent between 9 and 25 rest masses, a result consistent with later measurements of Voyvodic (105) on π -mesons and electrons, and of Daniel *et al.* (106) on cosmic ray particles. [See also Jauneau & Hug-Boussier (107) and Danysz, Lock & Yekutieli (108).] Other experiments on cosmic rays by Shapiro & Stiller (109), however, show an increase to a plateau at 12 to 14 per cent above minimum, reached only at energies as high as 100 rest masses. Several theoretical interpretations of the rise were given recently. Using Fermi's theory, Sternheimer (111) finds that only a small fraction of the Cerenkov radiation can escape from the silver bromide crystals, a result in contradiction with the calculations of Janssens & Huybrechts (100). Along similar lines, and taking into account the finite widths of the resonance frequencies of the medium, Budini's (112) results, depending sharply on the ratio between the width of the Cerenkov bands and the density of the medium, can be brought in good agreement with Morrish's (104) results. Huybrechts & Schönberg (113), taking more radical steps, show that in Fermi's method, the solution used for the fields at large distances depend quite strongly on the polarization of the medium in the immediate neighborhood of the track. They give a modification of Fermi's theory and their results also seem to be in agreement with Morrish's (104) observations and with the measurements of Gosh, Jones & Wilson (114) on the ionization of very fast particles in oxygen. In examining these results it is useful to remember, as indicated by Occhialini (101), that the relation of grain count to ionization also should be very closely studied experimentally in the region immediately before the minimum, and that part of the observed variation of grain density before and after the minimum may correspond to some mechanism specific to grain formation. The existence of the increase in principle extends the possibility of particle discrimination up to about 20 rest masses or more. The increase is however so small that reliability of measurements can only be obtained in very controlled conditions of image development and counting technique.

For low grain densities, grains only rarely cluster together and best reliability in counting is usually obtained by so called "blob counting" (i.e., counting each cluster as one grain regardless of shape). This can usually be done up to 1.5 times the minimum. When tracks of higher grain density are counted, this method obviously no longer can be used and some convention must be made on how to resolve clusters of irregular shapes into number

of grains. For instance, a number of grains is assigned to a group in proportion to its length (2). An interesting statistical study of grain counting is given by Hodgson (115) who shows that the statistical distribution of grains is slightly more peaked than a normal distribution because of finite grain size, an effect analogous to counting losses in Geiger-Müller tubes attributable to finite resolving time [see also (116)]. This apparent slight increase in the precision expected from grain counting usually cannot be exploited, because variations in grain count throughout the thickness or over the area of any single plate are difficult to avoid, even with the most refined developing techniques (35, 104, 109) and even more so between different plates.

For high grain densities it is sometimes useful (117, 118, 119) to measure the ratio of length occupied by gaps to the length of a fraction of track. This determination may be less easily affected by personal errors than grain counting.

A very promising recent development is the replacement of the eye by a photometric device for evaluation of dense ionization. Von Friesen & Kristiansson (120, 121) describe a simple photometric method where the image of the track is focussed upon a slit corresponding to a field about 1.5μ wide and 30μ long. The light passing through the slit falls on the cathode of a photomultiplier tube connected to a galvanometer. Readings are taken alternately on the track and on the background on both sides, and a value of the track blackening is obtained from their comparison. This quantity is found to be a reliable function of the energy loss for high ionizations (above 4 times minimum). While the photometric method is convenient for emulsions of thicknesses up to 200μ , it is more difficult to use for thick emulsions, because light coming from an object situated at the bottom of the emulsion is scattered and diffused in the top layers resulting in a marked variation of contrast with thickness. Rather important corrections must be made to compensate for this effect, an example of which will be found in (122). Variants of the methods are reported by several authors (123 to 125). Ceccarelli & Zorn (126) used a similar photometric arrangement to measure the width of the tracks. Using a narrower slit and moving it in a direction transverse to the track, they show that the shape of the blackening distribution across the track is practically unaffected by the amount of ionization, while the half-width of this distribution is a reliable function of ionization. The measurements are not sensitive to emulsion thickness. This variant of the photometric technique is more specially suited for the very high-energy loss of multiply charged particles.

Measurements of high ionization by grain counting can be successfully made over a limited range, selected by choosing emulsions of adequate low sensitivity and by precise control of the degree of underdevelopment. Experiments using this method and details of application cannot be discussed here, but the reader will find examples in experiments on the neutron disintegration of lithium (127, 128) and boron (35) and in studies of the heavy primaries of the cosmic ray radiation (14, 129). Recently, Ilford announced the production of plates with alternate layers of G 5 emulsion and of a new,

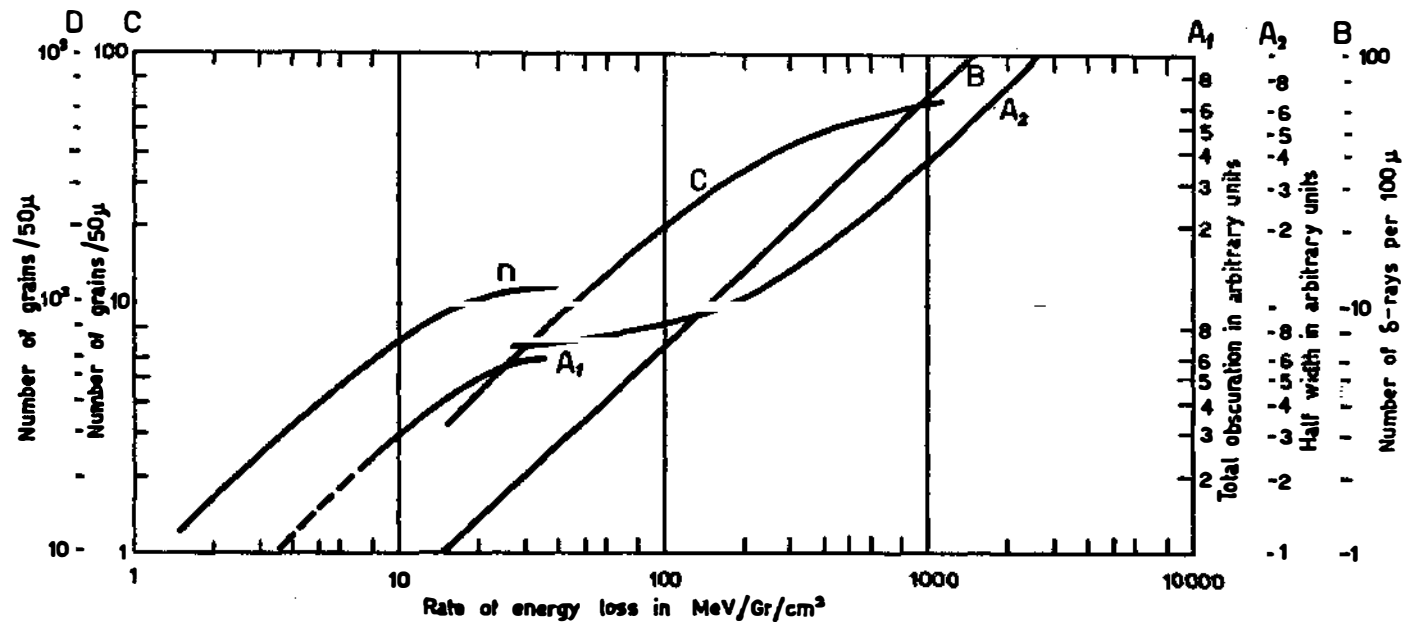


FIG. 3. Comparison of methods for the measurement of energy loss. From [Ceccarelli & Zorn (126). 'A₁'] Total obscuration, Ilford G 5. 'A₂'] Half-width, Ilford G 5. B. Density of δ-rays (more than 3 grains) Ilford G 5 from (131). C. Grain density in underdeveloped Kodak NTA from (129). D. Grain density in Ilford G 5 (149).

low sensitivity G 0 emulsion, adapted for use with processing methods usual for G 5 (20, 130). An indication of the ranges of applicability of the various methods for the measurement of ionization are given by Fig. 3.

Delta ray counting.—Particles of sufficiently high velocity or charge eject electrons from the atoms of the emulsion with energy high enough to make them appear as independent tracks originating usually at an angle of about 90° . The frequency of these δ -rays is discussed by Bradt & Peters (129) and, apart from a correction shown to be negligible for charges less than 30, is given by the formula: (when the atomic velocities of the electrons of the medium are neglected)

$$n = 2\pi N \left(\frac{e^2}{mc^2} \right)^2 \frac{z^2}{\beta^2} \left(\frac{mc^2}{E_1} - \frac{mc^2}{E_2} \right)$$

where n is the number of δ -rays per unit length of track, N is the number of electrons (of mass m) per unit volume of emulsion, and z and $\beta = v/c$ are the particle charge and velocity. E_1 and E_2 are, respectively, the lowest and highest electron energies that will produce observable tracks. For slow particles, E_2 is the maximum transferable energy $2mc^2\beta^2$. Emulsion sensitivity defines the value of E_1 , and of E_2 for fast particles, but not in a very critical way. In Ilford C 2 emulsion $E_1 = 10$ to 15 kev, depending on the convention used for electron track recognition (for instance, that they should have at least 4 grains) and $E_2 = 30$ kev. In Ilford G 5 emulsion, E_1 is again about 10 kev, and E_2 , mainly limited by the difficulty of recognizing electron tracks of low grain density and high scattering, is about 75 kev (129, 131). It is seen that as the δ -ray density depends on the square of the charge and on the particle velocity, it offers an interesting method of measurement for high charges, slow velocities, or both.

In early systematic use of δ -ray counting in the less sensitive Ilford C 2 emulsion, the observed number of δ -rays was a fraction only of the theoretical number, as they could only be observed when they were lying almost parallel to the plane of the emulsion. Careful investigations on the tracks of slow particles with Ilford G 5 emulsions showed very good agreement with theory (132). But for fast particles, recent observation of Dainton, Fowler & Kent (131) do not seem to be in agreement with the above formula. Comparing the δ -ray counts on slow protons and relativistic α -particles, they obtained evidence for the proportionality between δ -ray count and grain count. Their direct experimental evidence is limited to the region $\beta < 0.54$. In the opinion of these authors, however, indirect evidence for the validity of their law for higher values of β results from its application to a study of the presence of the lighter elements (Li, Be, B) in the multiply charged component of the cosmic ray radiation at high altitude. In contrast with the theoretical formula, their empirical relation resolves the charge spectrum into distinct peaks corresponding to these elements, in appreciable intensity. The existence of these light elements in cosmic rays is, however, the subject of contradictory results (14, 131), a controversy that cannot be examined here. It is clear that direct supporting experimental or theoretical results would be most useful in the high-energy region.

MULTIPLE SCATTERING

The theory of the measurement of scattering in the tracks of particles passing through nuclear emulsion comprises three somewhat distinct problems: the differential cross section for individual single scatterings, the statistical combination of the successive single scatterings into a multiple deflection probability function, and the analysis of a sample (a few measurements) in terms of the multiple scattering distribution function and of the experimental errors of measurement.

The potential of the nuclei responsible for scattering is not a pure Coulomb field. At large distances it is modified by the screening effect of the orbital electrons, at short distances the finite size of the nucleus (and the possibility of nuclear interaction) must be taken into account. The effect of screening was investigated in detail by Molière (133) with a Thomas-Fermi approximation of the atomic potential. His expression of the differential cross section for single scattering is valid for all velocities and charges, and agrees with the previous asymptotic formulae of Williams (134).

It is customary to measure the deflections projected on a plane, usually the plane of the emulsion itself. The combined effects of the large number of single scatterings suffered by a particle traversing a given thickness of material give rise, in first approximation, to a Gaussian distribution of projected deflections [Fermi (135)]. A better approximation [Williams (134)] takes into account the relatively high probability of large single scattering (more frequent than a number of small scatterings adding up to a large angle). It adds to a central Gaussian distribution a "tail" of relatively high probabilities for large angles. The exact distribution function is now available from the work of Molière (133), Snyder & Scott (136), Goudsmit & Saunderson (137), as applied by Berger (138) [see also Sauter & Wanke (139)]. Comparisons between these various treatments are found in (140 to 146). The effect of the finite size of the nucleus on the shape of the distribution was recently investigated by Olbert (145).

The quantity which is usually compared with experiments and called "the mean angle of scattering" is the arithmetic mean of the absolute value of the angles of deflection corresponding to a number of traversals of equal thicknesses of material. It has become customary to express the theoretical results in terms of the mean angle $\langle\alpha\rangle$ (in degrees) between successive chords to the track by the formula:

$$\langle\alpha\rangle = K(z/pv)(t/100)^{1/2}$$

where pv , the product of the particle momentum and velocity, is expressed in Mev, z , its charge, is in electron charges, t , the thickness of traversed material, is in microns, and K is called the scattering factor. The scattering factor K is a slowly varying function of the thickness t and, to a lesser degree, of the particle velocity v and charge z ; for instance, for $v=c$ and $t=100$, $K=25.8$ in Ilford G 5 emulsion (146). The various theories quoted give values of K which agree within 1 per cent, when Molière's expression for the single scattering differential cross section is inserted (140 to 146). It

is seen that the measurement of the mean angle of scattering gives a direct evaluation of z/pv . In contrast with other determinations that can be performed on the tracks, it gives information on the particle energy (or energy per nucleon) even in the relativistic region. Thus energies can be evaluated up to 10^{10} or 10^{11} ev with remarkable accuracy; provided long tracks are available (several mm.).

Ideally, the multiple scattering is determined by measuring the angles between successive tangents to the track at regular intervals. In the actual case, exact tangents cannot be drawn, as the track is defined only by a number of developed grains of finite size unevenly spaced and not necessarily centered on the exact trajectory. To overcome this difficulty, two experimental methods have been used, which can be termed the "angular" method and the "sagitta" or "coordinate" method. In the angular method (147) the average direction of the track over a certain segment a is determined by fitting a straight line as closely as possible to the centers of gravity of the grains in the segment. This operation is repeated at regular intervals t (usually called "cells"). Replacing the exact tangents by such average directions corresponds to a certain "smoothing" of the scattered angles, reducing the mean angle of scattering in a ratio depending on a/t . For the limit $a \ll t$ the best fitted line is close to a tangent and the smoothing factor is unity. For the other limit $a = t$ (angles between successive best fitted lines) Molière (148) has shown in the Gaussian approximation that the smoothing factor is $(26/35)^{1/2}$. Intermediate cases are also given by Molière. In the coordinate method (149) the track is lined along a straight reference line (one of the rectilinear motions of the microscope stage, say x). In principle, the distance between the line and the track is accurately measured at regular intervals. (Readings are taken of the coordinate y with a precise eye-piece micrometer.) It is easily shown that the second differences between the successive readings are proportional to the angles between successive chords of the track. In the Gaussian approximation, the mean angle between the successive chords is $(2/3)^{1/2}$ of that between tangents, but a more precise determination of mean chord angle is given by Scott (146), Berger (138), and Molière (133, 148).

Several types of errors can be recognized in the measurement. The finite length of the track limits the number of statistically independent readings (statistical error). The finite size of the grains and their distribution give a spurious scattering or "grain noise." Imperfections of the microscope stage may not allow a motion of the track strictly parallel to itself in the angular method, or on a perfect straight line in the sagitta method ("stage noise"). The readings are made with a certain personal error. Finally, emulsion distortion can appreciably influence the results. Much work has been devoted to the study and elimination of these various sources of error. Stages of high accuracy were designed (54), stage noise was determined by observation of almost straight tracks (150) or by optical and interferometric control (54). Personal errors in reading are eliminated by perfecting the reading instru-

ment (54) or by repeated measurements. Improvements in processing methods were aimed at reducing the distortion. Noise and true scattering are different functions of cell length and series of measurements with various cell lengths can be compared to separate true and spurious scattering (141, 151). While the angles between successive tangents are statistically independent, a correlation between successive measurements of these angles exists because of measuring errors (148, 152, 153). A statistical method can be used to evaluate the correlation of the experimental measurements and thus to eliminate the influence of the errors. The same procedure may be applied to the somewhat more elaborate case of sagitta measurements. The ratio between true and spurious scattering imposes the choice of the cell lengths which must increase with increasing particle velocity and momentum. Thus the number of cells drawn on a track of finite length is limited and fixes the value of the statistical error. It is tempting to try to augment the accuracy by using overlapping cells. This procedure has received much attention (148, 154, 155), as the successive readings are statistically correlated and their analysis poses interesting statistical problems; but it appears that the advantage of using the technique of overlap is rather small. More fruitful is the method of Mabroux (156) in which the angle of scattering projected on a plane perpendicular to the plane of the emulsion is measured by a precision control of the vertical motion of the objective: these measurements are completely independent of measurements in the plane of the emulsion.

The tail of the scattering distribution has an influence on the analysis of observations. For instance, the presence of one large angle among a set of readings may appreciably contribute to the observed mean angle. It is shown that the statistical error affecting the result can be reduced, if such large angles are eliminated in a consistent fashion. A common procedure is to eliminate by cut and try all observed angles larger than four times the mean. Other methods were also proposed (157). The theoretical value of the mean angle of a distribution cut-off at four times the mean, is given by numerous authors (146). It is found to be about 11 per cent below the mean angle of the full distribution, while about 1 per cent of the angles lie above the cut-off.

A number of investigations have been devoted to comparing theory and observations by measuring the scattering of particles of known energy. Monokinetic electrons were used by Corson (158), Menon, O'Ceallaigh & Rochat (157), and Voyvodic & Pickup (142), while the scattering of fast protons was calibrated by Gottstein & Mulvey (159) and Berger (138). Measurements were made by Bosley & Muirhead (160) in diluted G 5 emulsions. Most of the results are reviewed in (138, 141) and show a satisfactory agreement between theory and experiment. The theory of Molière has also received good experimental support by other methods (161). One measurement on slow electrons in photographic plates shows, however, an appreciable discrepancy [Hisdal (144)].

COMBINATION OF TECHNIQUES

Singly charged particles.—A particle producing a track in a nuclear emulsion must usually be identified as for mass, charge (possibly with sign) and energy. In principle, three independent measurements are needed to determine these three quantities. As the energy loss varies with the square of the charge, singly charged particles are easily recognized and two measurements ascertain their mass and energy. It is of course an essential feature of the nuclear emulsion method that some of the unstable particles can be recognized by their characteristic decay or interaction at the end of their range, but the discussion of these cases is outside the scope of the present review.

The combination range and ionization is of necessity limited to particles coming to rest in the emulsion. The variation of grain density (or gap density) with range was used, in early measurements of the mass ratio between π and μ mesons (162), where a detailed account of the method is found. The technique is still useful for the rare and interesting observations of heavy mesons (τ , χ , κ . . .) where it affords a determination of their mass (163). A photometric determination for such an event is found in (122). The measurement of scattering versus range is also limited to tracks stopped in the emulsion. It has the advantage over the previous combination of being more independent of the emulsion sensitivity and amount of development, but for short tracks it is affected by a larger statistical error. The measurement was also used in early determinations of meson masses (147) and is also useful for the analysis of heavy meson events (163). The method was put in a very convenient form by Menon & Rochat (164), who obtain a satisfactory

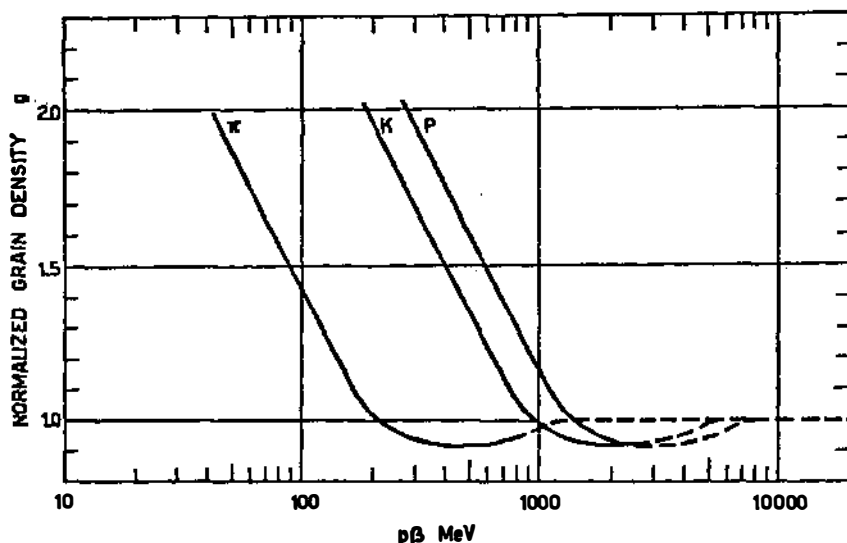


FIG. 4. The relation between grain density and $p\beta$ given by a measurement of scattering for singly charged particles. The dotted part of the curve shows where the rise of ionization after the minimum may affect particle identification (106).

discrimination between slow mesons, protons, deuterons, and tritons. When the minimum ionization sensitive emulsions first became available, it was soon recognized (101) that the combined measurements of grain count and scattering would be the most powerful means of investigating the many high-energy tracks occurring, for instance, in cosmic ray stars. A large fraction of our present knowledge of these events is based on such measurements [see for instance (13)]. The range of energies in which good discrimination is obtained between particles of different masses critically depends on the amount of increase in grain density after the minimum, discussed previously (Fig. 4). Many of the most interesting disintegrations observed in plates exposed to cosmic rays at high altitude involve particles having energies equal to several times their rest masses. Most of the improved methods of grain count and scattering measurements are aimed at a better investigation of these events. For lower energies, grain counting can be replaced by gap measurements in combination with multiple scattering (165).

Deviation of relatively slow particles in a magnetic field in air or in vacuum, and subsequent recording of their directions and ranges in photographic emulsions were used in numerous experiments [see for instance (4, 10)]. Thus the charge of the particle is readily determined, and very good accuracy is obtained for mass measurements. In the emulsion itself, the curvature of fast tracks by a strong magnetic field was successfully observed by

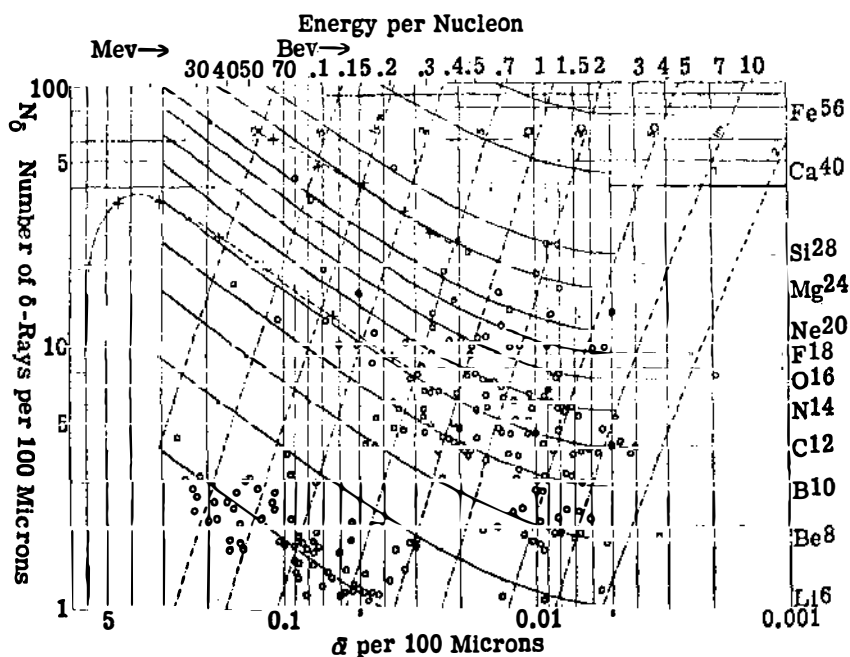


FIG. 5. The relation between δ -ray count, multiple scattering and range (dotted lines) for multiply charged particles. In calculating this chart Dainton, Fowler & Kent (131) used their empirical relation for δ -ray density.

Dilworth *et al.* (166). The relatively high multiple scattering masks the magnetic curvature unless very high fields and sufficiently long tracks are used. When the direction of motion of the observed particle is known, the sign of the charge is determined with a certain probability. For instance, with a field of 34,000 gauss, the sign will be determined correctly for about 80 per cent of the tracks of 5 mm. length and for 90 per cent of the tracks of 1 cm. Examples of the use of this technique for the study of high-energy cosmic ray events are given in (167, 168).

Multiply charged particles.—For multiply charged particles it has been possible in most experiments to use the approximation of the mass being nearly equal to twice the charge. Two independent measurements are again sufficient, in principle, to determine charge and energy.

For particles coming to rest in the emulsion, the charge can be deduced from the taper length, or from the characteristic increase in δ -ray density somewhat before the end of the range. Insensitive emulsions can be used to determine the ionization. Curves will be found in (14) giving the ionization and the δ -ray density as a function of the range and the charge. Discussions of the precision that can be obtained by these methods will be found in (14, 92). For particles in the relativistic range, a measurement of δ -ray density, or of ionization, determines uniquely the charge. The wider range of applicability of the δ -ray count can be seen in Fig. 3, but it is less accurate than the measurements of ionization in insensitive plates, which is probably preferable for $z < 6$. A measurement of multiple scattering gives a value of the energy per nucleon. Fig. 5 graph indicates the region where measurements of multiple scattering, δ -ray count, and range can be combined.

LITERATURE CITED

1. Rossi, B., *High-Energy Particles*, 127 (Prentice-Hall, Inc. New York 11, N. Y. 569 pp., 1952)
2. Fowler, P. H., and Perkins, D. H. *Fundamental Mechanism of Photographic Sensitivity*, 340 (Butterworths Scientific Publications, London, England, 345 pp. 1951)
3. Rotblat, J., *Progress in Nuclear Physics*, Frisch, O. R., Ed., 37 (Butterworth-Springer Ltd., London, 224 pp., 1950)
4. Beiser, A., *Revs. Mod. Phys.*, **24**, 273 (1952)
5. Vigneron, L., *J. phys. radium*, **14**, 121 (1953)
6. Shapiro, M. M., *Revs. Mod. Phys.*, **13**, 58 (1941)
7. Demers, P., *Can. J. Research*, [A]**25**, 223 (1947)
8. Powell, C. F., and Occhialini, G. P. S., *Nuclear Physics in Photographs* (Clarendon Press, Oxford, England, 1947)
9. Yagoda, H., *Radioactive Measurements with Nuclear Emulsions*, (John Wiley & Sons, Inc. New York 16, N. Y., 356 pp., 1949)
10. Powell, C. F., *Repts. Progr. in Phys.*, **13**, 350 (1950)
11. Waller, C., *The J. of Photographic Science*, **1**, 41 (1953)
12. McAllister, J. M., and Kean, D. A., *Proc. Phys. Soc. (London)*, [A]**64**, 91 (1951)
13. Camerini, U., Lock, W. O., and Perkins, D. H., *Progress in Cosmic Ray Physics*, **1**, Chap. 1 (North Holland Publishing Cy., Amsterdam, Holland 557 pp. 1951)

14. Peters, B., *Progress in Cosmic Ray Physics*, 1, Chap. 4 (North Holland Publishing Cy., Amsterdam, Holland, 557 pp., 1951)
15. *Kodak Nuclear Track Plates and Pellicles* (Eastman Kodak Company, Rochester, N. Y.)
16. *Kodak Nuclear Track Emulsions* (Kodak Limited, Research Laboratories, Wealdstone, Harrow, England, 1949)
17. *Ilford Nuclear Research Emulsions* (Ilford Research Laboratories, Ilford, London, England, 1949)
18. Demers, P. *Compt. rend.*, **231**, 616 (1950)
19. Jenny, L., *Fundamental Mechanisms of Photographic Sensitivity*, 259 (Butterworths Scientific Publications, London, England, 345 pp., 1951)
20. *Ilford Nuclear Research Emulsions, Suppl.* (Ilford Research Laboratories, Ilford, London, England, 1952)
21. Dodd, E. C., and Waller, C., *Fundamental Mechanisms of Photographic Sensitivity*, 266 (Butterworths Scientific Publications, London, England, 345 pp., 1951)
22. *Ilford Nuclear Research Emulsions in Gel Form* (Ilford Research Laboratories, Ilford, London, England, 1952)
23. Lord, J. J., *Phys. Rev.*, **81**, 901 (1951)
24. Dollman, E. M., *Rev. Sci. Instr.*, **21**, 118 (1950)
25. Goldschmidt-Clermont, Y., Osborne, L. S., and Scott, M. B., *Phys. Rev.*, **89**, 329 (1953)
26. Albouy, G., and Faraggi, H., *Rev. Sci. Instr.*, **22**, 532 (1951)
27. Goldstein, A. M., and Sherman, C. H., *Rev. Sci. Instr.*, **23**, 267 (1952)
28. Rotblat, J., and Tai, C. T., *Nature*, **164**, 835 (1949)
29. Vigneron, L., *J. phys. radium*, **10**, 305 (1949)
30. Coates, A. C., *Fundamental Mechanisms of Photographic Sensitivity*, 320 (Butterworths Scientific Publications, London, England, 345 pp., 1951)
31. Berriman, R. W. *Fundamental Mechanisms of Photographic Sensitivity*, 272, Butterworths Scientific Publications, London, England, 345 pp., 1951)
32. Beiser, A., *Rev. Sci. Instr.*, **23**, 500 (1952)
33. Summerfield, M. B., *Phys. Rev.*, **89**, 340 (1953)
34. Dilworth, C. C., Occhialini, G. P. S., and Payne, R. M., *Nature*, **162**, 102 (1948)
35. Dilworth, C. C., Occhialini, G. P. S., and Vermaesen, L., *Bull. centre phys. nucleaire univ. libre Bruxelles*, No. 13[a], 19 (1950); *Fundamental Mechanisms of Photographic Sensitivity*, 297 (Butterworths Scientific Publications, London, England, 345 pp., 1951)
36. Wilson, M. J. and Vanselow, S., *Phys. Rev.*, **75**, 1144 (1949)
37. Dainton, A. D., Gattiker, A. R., and Lock, W. O. *Phil. Mag.*, **42**, 396 (1951)
38. Yagoda, H., *Phys. Rev.*, **79**, 207 (1950); **80**, 753 (1950)
39. Herz, A. J., *J. Sci. Instr.*, **29**, 15 (1952); **29**, 60 (1952)
40. Stiller, B., Shapiro, M. M., and O'Dell, F. W., *Phys. Rev.*, **85**, 712 (1952)
41. Herz, A. J., and Edgar, M., *Proc. Phys. Soc. (London)*, [A]66, 115 (1953)
42. Marguin, G., *J. phys. radium*, **14**, 43 (1953)
43. Bonetti, A., Dilworth, C. C., and Occhialini, G. P. S., *Bull. centre phys. nucléaire univ. libre Bruxelles*, No. 13[b], 14 (1951)
44. Mignone, G., *Nuovo cimento*, **8**, 896 (1951)
45. Juric, M. K., and Smokovic, Z. A., *Rev. Sci. Instr.*, **23**, 564 (1952)
46. Stevens, G. W. W., *Nature*, **162**, 526 (1948)
47. Faraggi, H., Bonnet, A., Cohen, M. J., *J. phys. radium*, **13**, 105A (1952)
48. Gailloud, M., and Haenny, C., *Science et ind. phot.*, **23**, 221 (1952); *Mem. Soc. vaudoise des sci. nat.*, **10**, 271 (1952)

49. Dilworth, C. C., Occhialini, G. P. S., and Samuel, E., *Bull. centre phys. nucléaire univ. libre Bruxelles*, No. 22 (1948)
50. Powell, C. F., *Phil. Mag.*, **44**, 219 (1953)
51. Cosyns, M., and Vanderhaege, G., *Bull. centre phys. nucléaire univ. libre Bruxelles*, No. 15, 4 pp., (1950)
52. Major, J. V., *Brit. J. Appl. Phys.*, **3**, 309 (1952)
53. Kaplon, M. F., Peters, B., Reynolds, H. L., and Ritson, D. M., *Phys. Rev.*, **85**, 295 (1952)
54. Cosyns, M. G. E., *Bull. centre phys. nucléaire univ. libre Bruxelles*, No. 30, 13 (1951)
55. Barbour, I., *Rev. Sci. Instr.*, **20**, 530 (1949)
56. Putnam, T. M., and Miller, J. F., *Rev. Sci. Instr.*, **23**, 760–61 (1952)
57. Cosyns, M., and Vanderhaege, G., *Bull. centre phys. nucléaire univ. libre Bruxelles*, No. 12, 7 pp. (1950)
58. Bramson, H. J., Seifert, A. M., and Havens, W. W. Jr., *Phys. Rev.*, **88**, 304 (1952)
59. Occhialini, G. P. S., and Payne, R. M. (Unpublished data, 1947)
60. Masket, A. V., and Williams, L. B., *Rev. Sci. Instr.*, **22**, 113 (1951)
61. *Nucleonics*, **7**, 1, 81 (1950)
62. Blau, M., Rudin, R., and Lindenbaum, S., *Rev. Sci. Instr.*, **21**, 978 (1950)
63. Roberts, F., and Young, J. Z., *Nature*, **167**, 231 (1951); **169**, 963 (1952)
64. Hirschberg, D., (Private communication)^a
65. Livingston, M. S., and Bethe, H. A., *Revs. Modern. Phys.*, **9**, 263, (1937)
66. Bakker, C. J., and Segre, E., *Phys. Rev.*, **81**, 489 (1951)
67. Brown, L. M., *Phys. Rev.*, **79**, 297 (1950)
68. Bethe, H. A., Brown, L. M., and Walske, M. C., *Phys. Rev.*, **79**, 413 (1950)
69. Vigneron, L., *J. phys. radium*, **14**, 145 (1953)
70. Cüer, P., *Compt. rend.*, **233**, 1121 (1946)
71. Webb, J. H., *Phys. Rev.*, **74**, 511 (1948)
72. Wilkins, J. J., *Atomic Energy Establishment (Harwell) Report, G/R 664* (1951)
73. Lattes, C. M. G., Fowler, P. H., and Cüer, P., *Proc. Phys. Soc. (London)*, [A] **59**, 883 (1947)
74. Rotblat, J., *Nature*, **165**, (1950), **167**, 550 (1951)
75. Catala, J., and Gibson, W. M., *Nature*, **167**, 551 (1951)
76. Nerenson, N., and Reines, F., *Rev. Sci. Instr.* **21**, 534 (1950)
77. Cüer, P., and Jung, J. J., *J. phys. radium*, **S. 12**, 52 (1951); *Science et ind. phot.*, **22**, 401 (1951)
78. Faraggi, H., *Compt. rend.*, **230**, 1389 (1950)
79. Panofsky, W. K. H., and Fillmore, F. L., *Phys. Rev.*, **79**, 57 (1950)
80. Bradner, H., Smith, F. M., Barkas, W. H., and Bishop, A. S., *Phys. Rev.*, **77**, 462 (1950)
81. Richards, H. T., Johnson, V. R., Ajzenberg, F., and Laubenstein, M. J. W., *Phys. Rev.*, **83**, 994 (1951)
82. Gailar, O., Seidlitz, L., Bleuler, E., and Tendam, D. J., *Rev. Sci. Instr.* **24**, 126 (1953)
83. Lees, C. F., Morrison, G. C., and Roser, W. G. V., *Proc. phys. Soc. (London)*, [A] **66**, 13 (1953)
84. Gottstein, K., and Mulvey, J. H., *Phil. Mag.*, **42**, 1089 (1951)
85. Ross, M. A. S., and Zajac, B., *Nature*, **162**, 923 (1948)
86. Herz, R. H., *Phys. Rev.*, **75**, 478 (1949)

^a Permission to mention work in progress is gratefully acknowledged.

87. Blum, J., *Compt. rend.*, **228**, 918 (1949)
88. Barkas, W. H., *Phys. Rev.*, **89**, 1019 (1953)
89. Longchamp, J. P., *J. phys. radium*, **14**, 89 (1953)
90. Perkins, D. H., *Proc. Roy. Soc. (London)*, [A]**203**, 339 (1950)
91. Freier, P., Logfren, E. J., Ney, E. P., and Oppenheimer, F., *Phys. Rev.*, **74**, 1818 (1948)
92. Hoang, T. F., *J. phys. radium*, **12**, 739 (1951)
93. Cüer, P., and Longchamp, J. P., *Compt. rend.*, **236**, 70 (1953)
94. Fermi, E., *Phys. Rev.*, **57**, 485 (1940)
95. Wick, G. C., *Nuovo cimento*, **1**, 302, (1943)
96. Halpern, O., and Hall, H., *Phys. Rev.*, **73**, 477 (1948)
97. Bohr, A., *Kgl. Danske Videnskab. Selskab Mat-fys. Medd.*, **24**, No. 19 (1948)
98. Messel, H., and Ritson, D. M., *Phil. Mag.*, **41**, 1129 (1950)
99. Schönberg, M., *Nuovo cimento*, **8**, 159 (1951)
100. Janssens, P., and Huybrechts, M., *Bull. centre phys. nucleaire univ. libre Bruxelles*, No. 27, 32 pp. (1951)
101. Occhialini, G. P. S., *Nuovo cimento Suppl.*, **6**, 377 (1949)
102. Bradt, H. L., Kaplon, M. F., and Peters, B., *Helv. Phys. Acta*, **23**, 24 (1950)
103. Pickup, E., and Voyvodic, L., *Phys. Rev.*, **80**, 89 (1950)
104. Morrish, A. H., *Phil. Mag.*, **43**, 533 (1952)
105. Voyvodic, L., *Report to the Bristol Conference on Heavy Mesons* (1951)
106. Daniel, R. R., Davies, J. H., Mulvey, J. H., and Perkins, D. H., *Phil. Mag.*, **43**, 753 (1952)
107. Jauneau, L., and Hug-Bousser, F., *J. phys. radium*, **13**, 465 (1952)
108. Danyasz, M., Lock, W. O. and Yekutieli, G., *Nature*, **169**, 364 (1952)
109. Shapiro, M. M., and Stiller, B., *Phys. Rev.*, **87**, 682 (1952); *Proceedings of the Third Annual Rochester Conference on High Energy Physics* (Interscience Publishers Inc. New York 3, N. Y., 120 pp, 1953)
110. Sternheimer, R. M., *Phys. Rev.*, **88**, 851 (1952)
111. Sternheimer, R. M., *Phys. Rev.*, **89**, 1148 (1953)
112. Budini, P., *Phys. Rev.*, **89**, 1147 (1953); *Nuovo cimento*, **10**, 236 (1953)
113. Huybrechts, M., and Schönberg, M., *Nuovo cimento*, **9**, 764 (1952)
114. Gosh, S. G., Jones, G. M. D. B., and Wilson, J. G., *Proc. Phys. Soc. (London)*, [A]**65**, 68 (1952)
115. Hodgson, P. E., *Brit. J. Appl. Phys.*, **3**, 11 (1952)
116. Happ, W. W., Hull, T., and Morrish, A. H., *Can. J. Phys.*, **30**, 669 (1952)
117. Hodgson, P. E., *Phil. Mag.*, **41**, 725 (1950)
118. Renardier, M., Moucharafieh, H., and Morand, M., *Compt. rend.*, **231**, 848 (1950)
119. Renardier, M., and Avignon, Y., *Compt. rend.*, **233**, 393 (1951)
120. von Friesen, S., and Kristiansson, K., *Nature*, **166**, 686 (1950); **168**, 377 (1951); *Arkiv. Fysik*, **4**, 505 (1952)
121. *Manne Siegbahn*, 623 (Almquist and Wiskells, Upsala, Sweden, 845 pp., 1951)
122. Kristiansson, K., *Phil. Mag.*, **44**, 268 (1952)
123. Kayas, G., and Morellet, O., *Compt. rend.*, **234**, 1359 (1952)
124. Della Corte, M., and Ramat, M., *Nuovo cimento*, **9**, 605 (1952)
125. Mathieu, R., and Demers, P., *Can. J. Physics*, **31**, 97 (1953)
126. Ceccarelli, M., and Zorn, G. T., *Phil. Mag.*, **43**, 356 (1952)
127. Mortier, M., and Vermaesen, L., *Bull. centre phys. nucleaire univ. libre Bruxelles*, No. 5, 6 pp. (1948)
128. Faraggi, H., *Compt. rend.*, **227**, 527 (1948)

129. Bradt, H. L., and Peters, B., *Phys. Rev.*, **80**, 943 (1950); **74**, 1828 (1948)
130. Herz, A. J., and Waller, C., *Phil. Mag.*, **43**, 593 (1952)
131. Dainton, A. D., Fowler, P. H., and Kent, D. W., *Phil. Mag.*, **42**, 317 (1951); **43**, 729 (1952)
132. Voyvodic, L., *Can. J. Research*, [A]**28**, 315 (1950)
133. Molière, G., *Z., Naturforsch.*, **2**[a], 133 (1947); **3**[a], 78 (1948)
134. Williams, E. J., *Proc. Roy. Soc. (London)*, [A]**169**, 531 (1938), *Phys. Rev.*, **58**, 292 (1940)
135. Fermi, E., in Rossi, B., and Greisen, K., *Revs. Mod. Phys.* **13**, 240 (1941)
136. Snyder, H. S., and Scott, W. T., *Phys. Rev.*, **76**, 220 (1949)
137. Goudsmit, S., and Saunderson, J. L., *Phys. Rev.*, **58**, 36 (1940)
138. Berger, M. J., *Phys. Rev.*, **88**, 58 (1952)
139. Sauter, F., and Wanke, H., *Z. Naturforsch.*, **7**[a], 33, (1952)
140. Goldschmidt-Clermont, Y., *Nuovo cimento*, **7**, 331 (1950)
141. Gottstein, K., Menon, M. G. K., Mulvey, J. H., O'Ceallaigh, C., and Rochat, O., *Phil. Mag.*, **42**, 708 (1951)
142. Voyvodic, L., and Pickup, E., *Phys. Rev.*, **85**, 91 (1952)
143. Molière, G., and Sauter, F., *Z. Naturforsch.*, **7**[a], 280 (1952)
144. Hisdal, E., *Phil. Mag.*, **43**, 790 (1952)
145. Olbert, S., *Phys. Rev.*, **87**, 319 (1952)
146. Scott, W. T., *Phys. Rev.*, **85**, 245 (1952)
147. Goldschmidt-Clermont, Y., King, D. T., Muirhead, H., and Ritson, D. M., *Proc. Phys. Soc. (London)*, **61**, 138 (1948)
148. Molière, G. (Unpublished data)^a
149. Fowler, P. H., *Phil. Mag.*, **41**, 169 (1950)
150. Levi-Setti, R., *Nuovo cimento*, **7**, 331 (1951)
151. Mabboux, C., *Compt. rend.*, **232**, 2309 (1951)
152. Moyal, J. E., *Phil. Mag.*, **41**, 1058 (1950)
153. d'Espagnat, B., *J. phys. radium*, **13**, 74 (1952)
154. O'Ceallaigh, C., and Rochat, O., *Phil. Mag.*, **42**, 1050 (1951)
155. Schonland, D. S., *Proc. Phys. Soc. (London)*, [A]**65**, 640 (1952)
156. Mabroux, C., *Compt. rend.*, **232**, 1091 (1951)
157. Menon, M. G. K., O'Ceallaigh, C., and Rochat, O., *Phil. Mag.*, **42**, 932 (1951)
158. Corson, D. R., *Phys. Rev.*, **80**, 303 (1950)
159. Gottstein, K., and Mulvey, J. H., *Phil. Mag.*, **42**, 1089 (1951)
160. Bosley, W., and Muirhead, H., *Phil. Mag.*, **43**, 63 (1952)
161. Hanson, A. O., Lanzl, L. H., Lyman, E. M., and Scott, M. B., *Phys. Rev.*, **84**, 634 (1951)
162. Lattes, C. M. G., Occhialini, G. P. S., and Powell, C. F., *Nature*, **453**, 486 (1947); *Proc. Phys. Soc. (London)*, **61**, 173, (1948)
163. O'Ceallaigh, C., *Phil. Mag.*, **42**, 1032 (1951)
164. Menon, M. G. K., and Rochat, O., *Phil. Mag.*, **42**, 1232 (1951)
165. Belliboni, G., and Merlin, M., *Nuovo cimento*, **8**, 349 (1951)
166. Dilworth, C. C., Goldsack, S. J., Goldschmidt-Clermont, Y., and Levy, F., *Phil. Mag.*, **41**, 1032 (1950)
167. Goldsack, S. J., and Page, N., *Phil. Mag.*, **42**, 570 (1951)
168. Demeur, M., Dilworth, C. C., and Schönberg, M., *Nuovo cimento*, **9**, 92 (1952)

^a Communication of these results is gratefully acknowledged.