

Tracks of Nuclear Particles in Photographic Emulsions

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I. EARLY HISTORY OF THE DIRECT PHOTOGRAPHIC METHOD

AN alpha-particle striking a photographic plate at glancing incidence alters the grains of silver bromide in its path. After development of the plate, microscopic investigation reveals each alpha-track as a minute trail of discrete silver grains. The discovery of this effect by M. Reinganum (R1)¹ in 1911 probably stemmed from the researches of O. Mügge on pleochroic halos, from the photographic investigations of S. Kinoshita, and from certain cloud-chamber experiments of C. T. R. Wilson.

In 1909, while studying the radioactive properties of various minerals, Mügge (M6) strewed some tiny crystals of zircon over a moist photographic plate. After twenty-five days the plate was developed and examined under a microscope. Black spots were observed where the zircon particles had fallen, and emerging from these spots were rows of black dots. Mügge correctly attributed this phenomenon to the radioactivity of the mineral. He did not, however, identify the radiation producing the series of dots as alpha-particles.

Working in Rutherford's laboratory in 1910, Kinoshita (K1) showed that a halide grain is rendered developable when struck by a single alpha-particle. This observation was based upon photometric density studies, and upon microscopic counts of the number of silver grains on a

plate previously exposed to a suitable radioactive source. Kinoshita suggested that the photographic action of an alpha-particle is due to the ionization of silver halide molecules in the grains. That Kinoshita failed to detect tracks as such may be ascribed to the fact that in his experiments alpha-rays were incident upon the plate at small angles with the normal, rather than at glancing angles. The projection of each track, viewed microscopically, would thus appear as a single dot.

Reinganum's attempt to produce such tracks in a photographic emulsion may have been inspired by a discovery of C. T. R. Wilson (W13). Early in 1911 Wilson had succeeded in making visible by photographic means the paths of single alpha-particles in his cloud chamber.

In 1912 W. Michl (M5) confirmed and extended Reinganum's findings. He measured the track length and number of grains per track for alpha-particles of different range, and concluded that both quantities are linear functions of the air range of the alphas used. The average track of a polonium alpha-particle, consisting of eight grains, was found to be 23 microns long. From this value the range in air was computed to be 3.8 cm, in good agreement with the Geiger value, 3.77 cm. Reinganum had reported a number of deflections in his tracks which he had attributed to scattering. Michl contended that most of these bent or curved tracks—which were more numerous near the edges of his own plates—can be explained by the particular way in which the emulsion contracts upon drying.

¹ A letter and a number, e.g. (R1), refer to an original paper. A list of references is given at the end of this monograph.

Using Kinoshita's method—in which single grains rather than tracks are registered—F. Mayer (M3) showed that the scattering of alpha-particles by aluminum foils may be demonstrated photographically by counting the number of grains in various areal strips of a plate. In 1914 W. Makower and H. P. Walmsley (M1) published the first dark-field photomicrographs showing the tracks of alpha-particles in a photographic emulsion. They pointed out that apparent deflections in tracks cannot, in general, be ascribed with certainty to scattering. However, by focusing the microscope on various horizontal planes in the emulsion, one can usually distinguish between a "bend" due to two contiguous tracks, and a real deflection in a single track. One of their pictures illustrates what the authors regarded as an authentic case of scattering.

Kinoshita and Ikeuti (K2), in 1915, "contaminated" an emulsion by touching it at a number of points with a needle having some radium C deposit at its tip. After storing the plate for some time and developing it, they noticed a number of minute dark spots. Magnified several hundred diameters, these spots resembled the pleochroic halos found in certain radioactive rocks. Each "halo" consisted of numerous alpha-tracks emerging radially from a "contamination center" and terminating on or near a sphere whose radius equalled the range of the alpha-rays in the emulsion (see Fig. 1, (B) and (C)). In the early experiments halos were obtained only for radium C. Subsequently, however, Ikeuti (I1) succeeded in registering the shorter radius halos of radium A as well. The ratio of this radius to that of the halos from radium C agreed closely with the ratio of the corresponding alpha-particle ranges in air.

E. Mühlestein (M7) introduced a further refinement in the halo technique by immersing the plate in mercury after it was "infected" and leaving it there until development. In the resulting halos, alpha-tracks issued principally from centers within the photographic emulsion, and only rarely from the tiny masses of radioactive dust lying over the surface of the emulsion. Consequently, the dark nuclei of the halos were greatly reduced in size, enabling a more accurate determination of the alpha-range. For radium C,

a track length of 50.0 microns was obtained in this way, and for polonium, 27.7 microns.

To test the effects of a magnetic field, Kinoshita and Ikeuti (K2) infected a plate with an active needle, then quickly placed it between the poles of a powerful electromagnet, with the plate surface perpendicular to the lines of force. No indications of curvature appeared in the tracks, although field strengths as high as 10,000 gauss were applied. This result was accounted for by a calculation of the curvature expected at the maximum field strength for alpha-particles moving with a velocity of 2×10^9 cm/sec. Since the radius of curvature under these conditions is about 40 cm, whereas the path in the emulsion is about 0.054 mm long, it is not surprising that any curvature which may have been present could not be detected.

With the needle-point technique of Kinoshita and Ikeuti, R. R. Sahni (S1) investigated the photographic effect of beta-rays, with results which differed strikingly from those for alpha-particles. Instead of showing approximate spherical symmetry, the "contamination nuclei" were irregular and nebulous. Moreover, the beta-particles left no straight tracks. Experiments with gamma-ray sources yielded spots similar to those made by beta-rays. This was attributed to the production of secondary beta-particles by the gamma-rays.

In another experiment, Sahni (S2) studied the scattering of alpha-particles by gases. From a needle tip activated with radium C, alpha-particles were allowed to pass through a small opening into a chamber containing the gas scatterer. After traversing the gas, the particles struck a photographic plate at the opposite end of the chamber. From the distribution of silver grains in concentric areas on the plate, it was found that the radial density gradient depends upon the pressure of the gas. The results were inadequate, however, for an accurate measurement of the most probable scattering angle.

That protons can produce tracks in photographic emulsions similar to those of alpha-particles was demonstrated by M. Blau (B5, B6) in 1925. Alpha-rays from a polonium source were directed at a thin layer of paraffin, from which protons were ejected. To absorb the alpha-particles which traversed the paraffin, a thin

copper foil was interposed between it and the photographic plate. After prolonged exposures tracks were also registered when the paraffin was replaced by aluminum. Subsequently, bombardment of carbon and beryllium with polonium alphas yielded proton tracks, which were ascribed to the disintegration of these elements (B7, B8).

This interpretation was based upon the earlier work of Kirsch and Pettersson (K3), who had studied such disintegrations with the scintillation method.

Blau compared the average distance between adjacent silver grains in proton tracks with those in alpha-tracks, and found the spacing to

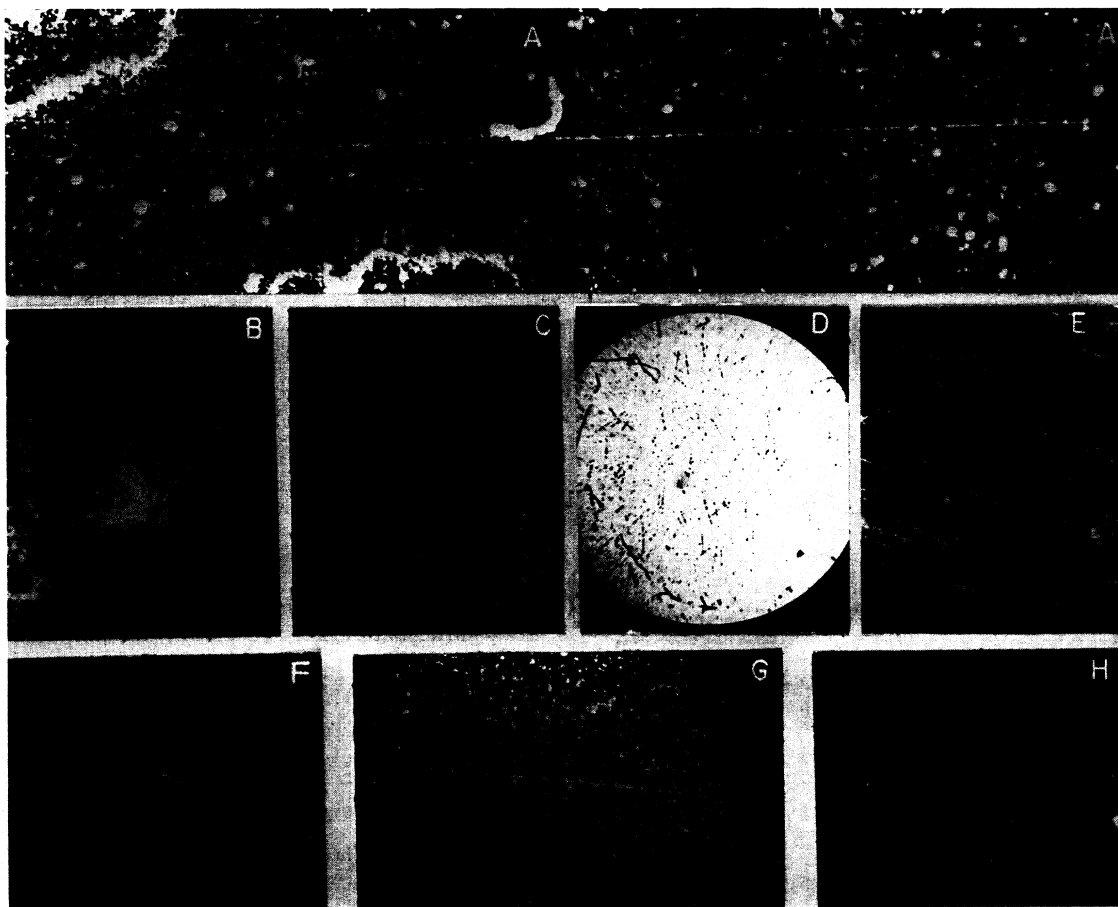


FIG. 1. (A) Cosmic-ray track from plate sent up in stratosphere balloon flight by T. R. Wilkins. Length of track in photographic emulsion, 0.55 mm. Interpreted from the grain spacing as a very high energy alpha-particle, whose range in air is about 75 cm. Magnification 260X.

(B) and (C) Alpha-rays emanating from "contamination centers" in an emulsion impregnated with polonium. Black streak in (C) is a fiducial mark in the microscope eyepiece.

(D) and (E) Po- α tracks obtained by exposing plate edgewise to a radioactive source. Thick black marks in (D) are caused by dust particles on the plate surface. A comparison of photographs (B) and (E) with (C) and (D) gives some indication of the advantage of dark-field illumination over bright-field. Of the latter pair, (C) is from an ordinary process plate; it shows a high background of fog; (D) was obtained with an Eastman Fine Grain Alpha-Particle plate.

(F) Fivefold fork due to successive disintegrations of a single radioactive atom or to alpha-particle emission from several contiguous atoms. At the lower left a short track is superposed over a longer one.

(G) Cosmic-ray track of an alpha-particle from plate exposed at an elevation of 6700 feet. Length in emulsion, 157 microns; range in air, about 22 cm.

(H) Threefold nuclear disintegration probably due to cosmic ray, from Ilford plate sent aloft into the stratosphere. To render the three tracks simultaneously visible although they do not lie in same plane, low magnification is used. The vertical track appears to be due to a proton; the horizontal one is an alpha-track. (Photographs (B), (E), and (F) are reproduced through the courtesy of Dr. Niel F. Beardsley.)

be greater in the former. This difference was especially pronounced in the case of fast protons, for which the average intergrain spacing was almost twice that for typical alpha-tracks.

In 1927 Myssowsky and Tschishow (M8) pointed out that the use of thickly coated plates would make it possible to register longer tracks with a wider range of angles of incidence. They prepared emulsions thicker than 50 microns, and published stereoscopic photomicrographs of deflections and forked tracks observed on such plates.

An important contribution by Wambacher and Blau (W1, B11) was their discovery that pinacryptol yellow sensitizes photographic emulsions to energetic protons. Curiously enough, the same organic dye is an effective desensitizer to visible light. Although protons liberated by the impact of alpha-particles could be recorded on ordinary plates, the recoil protons which result from neutron impacts were not registered unless the plate was first bathed in a solution of pinacryptol yellow. Since the dye rapidly loses its sensitizing property by oxidation, it was found necessary to expose the pretreated plates in a vacuum or in a nitrogen-filled container. On such plates, tracks were observed which indicated the existence of higher neutron energies than those hitherto reported. The pinacryptol yellow technique was further studied by Zila (Z1), and applied by Rumbaugh and Locher (R4) to cosmic-ray research.

To study the photographic process of proton detection, Blau and Wambacher (B13, B17) measured the density of silver grains in proton tracks as a function of the ionizing power of the rays. For protons with an air equivalent range of 2 cm or less, the grain density was found to be independent of the range, as it is also for alpha-particles. At the same time these workers extended their investigation of the sensitizing action of a number of organic and inorganic solutions. Dyes similar to pinacryptol yellow were found to be effective. The sensitizing property of these substances, according to the authors, is unrelated to their desensitization of plates to light. The dye does not, moreover, act as a bromide acceptor. Its adsorption seems to alter the outer surface of the silver bromide grain,

so that it is more readily affected by the impact of a proton.

Photographic emulsions sensitive to energetic protons without preliminary immersion in pinacryptol yellow were independently developed by A. Jdanov (J1) and by the Ilford laboratories (I2, T4). Jdanov showed that the nature of the tracks produced by nuclear particles depends on the size of the grain and on the concentration of silver bromide in the emulsion. Thus, to obtain good proton tracks, the grain diameter should be about 0.5–0.8 microns.

The development of proton-sensitive plates extended the applicability of the photographic method to problems in nuclear physics, and provided a new tool for the investigation of cosmic rays. Some of the recent advances in these fields will be reviewed in Sections III and IV. First, however, we shall examine more closely the nature of the photographic technique, its possibilities, and its limitations.

II. NATURE OF THE PHOTOGRAPHIC TECHNIQUE —ITS ADVANTAGES AND LIMITATIONS

The continuous sensitivity of the photographic plate is its basic advantage. In nuclear reactions with a low yield, the time of exposure may be conveniently extended as long as necessary to get sufficient data. Similarly, in the registration of infrequent cosmic-ray processes, the emulsion acts as a sort of continuously sensitive cloud chamber. A great deal of information can be accumulated on a single small plate; this permanent record can be subsequently examined at one's leisure. Because of their light weight, photographic plates can be sent up into the stratosphere by means of unmanned balloons; and since they need practically no attention during exposure, they may be left on high mountains for the prolonged periods required to record rare cosmic-ray events. While the actual microscopic examination of the plates may be very tedious, the method is essentially simple in principle.

On the other hand, the photographic method is subject to severe limitations. The most important of these are due to "fog." Examination of an unexposed plate under the microscope after development reveals an enormous number of silver grains—as many as hundreds of thousands

in one square centimeter of plate area. Certain precautions can be taken to reduce this background—e.g., exclusion of red light, filtration of processing solutions and washwater, avoidance of marked temperature variations. Regardless of the most painstaking care in development, however, a great number of grains appear.

This “unexposed” background may arise in many ways. Chemical action by the various processing solutions in which the plate is immersed is probably the chief fogging agent. During the history of the plate prior to development, excessive fluctuations in temperature, gamma-rays from radioactive materials in the surroundings, weakly phosphorescent materials in the dark-room—all these are capable of rendering silver bromide grains developable. Cosmic rays which do not produce tracks may, nevertheless, possess sufficient ionizing power to alter single grains. Even mechanical shock has been suspected of affecting the sensitive emulsion.

The presence of a heavy background of silver grains makes the task of finding and studying the tracks of nuclear particles very difficult. These tracks are typically a fraction of a millimeter in length, since the stopping power of the emulsion is about fourteen hundred times that of air. Moreover, each track consists of a row of discrete silver grains whose average distance apart is several times their own diameter. A stray silver particle near the beginning or end of a track will increase its apparent length. On the other hand, a nuclear particle may have continued for several microns after its encounter with the last visible silver grain, giving a measured range which is too short. Thus there is an uncertainty regarding the true terminals of a track.

Besides the effect of straggling in the emulsion, there are other sources of error. For particles which enter the emulsion at steep angles rather than at grazing incidence, determination of track length depends partly on measurement of depth. This is done by differential focusing, and is less accurate than the measurement of a track's horizontal projection with the eye-piece scale. The grain-spacing of a track increases as the temperature falls (S5). In experiments in which it is difficult to maintain temperature control—for example, during prolonged exposure of cosmic-ray plates—this effect introduces another uncer-

tainty. Finally, there are various spurious appearances many of which may be easily mistaken for real tracks. These may be produced by scratches, by contractions of the gelatine during processing, and by the presence of foreign matter in the emulsion. Certain criteria, however, are of help in distinguishing pseudo-tracks from real ones. One aid is the fact that scratches usually lie entirely on the surface of the emulsion.

In the microscopic examination of the plate, dark-field illumination is vastly superior to the usual bright-field. (See Fig. 1D and E.) Because it affords greater contrast, the tracks stand out much more clearly. Such illumination, unfortunately, also enhances the visibility of the many stray silver grains. Consequently, even under the most favorable conditions there is a considerable strain on the observer, and some error creeps in on this account. Another source of inaccuracy in the measurement of tracks is the finite size of the grains. For fairly large grains the center of a grain may be appreciably displaced from the true path of the nuclear particle.

To overcome some of these difficulties, the Ilford laboratories produced a fine-grained emulsion with greatly reduced stray background (I2). This so-called “R1-plate” was insensitive to protons. Further trials yielded an emulsion—the “R2-plate”—sensitive to protons, but somewhat less free from stray silver grains. Subsequently, Ilford developed the so-called “New Halftone” emulsion, which is even more sensitive to energetic protons.

Using the R2 emulsion, Taylor (T4) was able to distinguish roughly between the tracks of protons and alpha-particles by measuring the average spacing (\bar{s}) between their grains. He plotted the frequency of the two kinds of tracks against different values of \bar{s} . From this frequency distribution he concluded that an unknown track with \bar{s} greater than two microns is probably due to a proton; if \bar{s} is less than two microns, it is probably produced by an alpha-particle. In general, it may be said that identification is possible when dealing with large numbers of particles, but much less reliable for single events.

The Agfa laboratories developed an emulsion—the “K-plate”—which is also relatively free from background. According to Wambacher (W3), this emulsion shows a greater difference between

protons and alpha-tracks than the Ilford plates. The Eastman Kodak laboratories, collaborating with T. R. Wilkins, have also produced a plate suitable for the registration of nuclear particles—the so-called “Fine Grain Alpha-Particle Plate.”

Wilkins and St. Helens (W11) made a careful study of the relative grain-spacing of alpha, proton, and deuteron tracks in the R2 emulsion. Their proton and deuteron beams were obtained from a cyclotron. Plotting the total number of grains per track against the track length, they obtained a distribution of three bands of points corresponding to the three kinds of particles, the deuteron band lying between the other two. Wilkins (S5) suggested a theory of variation in grain spacing depending upon the fact that the photographic action of corpuscular radiations is due to ionization. He applied Bloch's formula for the energy loss of a high speed particle of charge e and velocity V . In traversing a medium of atomic number Z , such a particle loses kinetic energy T at the rate

$$-\frac{dT}{dx} = \frac{4\pi e^4 N Z^2}{m V^2} \log \frac{4\pi m V^2}{k Z R h},$$

where m is the electronic mass, R the Rydberg constant, N the Loschmidt number and k an undetermined constant. From his theory, Wilkins deduced a ratio of 2 : 1 for the average grain spacing produced by protons and alpha-tracks, and 1.25 : 1 for protons and deuterons. The corresponding observed ratios were about 1.8 and 1.4, respectively.

The relative insensitivity of photographic plates to electrons imposes a restriction upon their applicability. In cosmic-ray investigations, however, this restriction must be regarded as advantageous; for if energetic electrons could leave tracks in plates, their relative abundance would make it difficult or impossible to study the rare tracks due to heavy particles. An exposure sufficiently long to register an appreciable number of cosmic-ray protons would leave the plate thoroughly fogged by electron tracks. Thanks to the selective sensitivity of the plate to heavy nuclear particles, the photographic method is making a distinctive contribution to research in cosmic radiation.

Because of the many sources of error which arise in microscopic inspection, it is important to employ as good a viewing technique as possible. The advantage of using a dark-field condenser has been described above. In addition, the observer will find a binocular microscope of great value in reducing eye-strain and fatigue. A graduated mechanical stage is essential, while an eyepiece scale is useful even in a qualitative investigation. Hyperplane eyepieces provide a flatter image plane than Huygenian eyepieces, the difference being conspicuous at the periphery of the field. Wilkins (W11) recommends the use of a variable iris diaphragm in the tube of the objective for convenient adjustment of the illumination and depth of field. Stereoscopic photomicrography has been applied to measurement of track lengths and angles between tracks (J1, M2, M8).

A review of the photographic emulsion technique would be incomplete without reference to the investigations of the nature of the latent image produced by corpuscular radiation (B10, B12, E1, L1). Since these are concerned with the photographic process itself rather than its application to nuclear physics, they will not be described here. (For further general discussions of the photographic method, see references B22, B23, P1, T11, W3 and W12.)

III. CONTRIBUTIONS OF THE PHOTOGRAPHIC METHOD IN THE FIELD OF COSMIC RAYS

The occasional occurrence in cloud chambers of tracks due to heavy² cosmic-ray particles suggested that the photographic emulsion might be a suitable means of recording them, especially at high altitudes. In the stratosphere flight of the balloon “Explorer II,” conducted by the National Geographic Society and the U. S. Army Corps in 1936, Wilkins (W5, W6, W7) and Rumbaugh and Locher (R4, R5) sent along some Ilford R2 plates in the gondola.

In addition to the alpha-tracks left by radioactive impurities, Wilkins found a number of longer tracks which he believed were produced by alpha-particles and protons of cosmic-ray

² The word “heavy” will be used here to describe particles at least as massive as protons.

origin. Of special interest was a very long horizontal track containing about 350 grains, whose spacing indicated that it was produced by an alpha-particle. (See Fig. 1(A).) At several points along the path, there emerged shorter single tracks, apparently produced by ejected protons. At such points the main track was slightly bent. The total energy of the event was estimated to be about 100 Mev. Takeuchi and his associates (T1) found a similar track in a plate exposed at the summit of Mt. Huzi, 3776 m above sea level. Wilkins also observed a number of forked tracks which he interpreted as the effects of nuclear disintegrations.

Rumbaugh and Locher covered some of their plates with paraffin, to obtain evidence about cosmic-ray neutrons; and others with materials representing various elements, to test the possibility that the cosmic rays eject heavy nuclear particles in their reaction with these elements. In the paraffin-covered emulsions, 4.5 ± 1 proton tracks per sq. cm were found, whereas less than one per sq. cm occurred in plates covered with carbon and lead. The protons were regarded as recoils from neutron collisions. Similarly covered control-plates left on Pikes Peak for one month showed even fewer proton tracks than those in the stratosphere emulsions. It was concluded that at a depth of one-half meter water equivalent below the top of the atmosphere, neutrons constitute a considerable fraction of the cosmic radiation in number, though not in energy. In plates covered with aluminum, small numbers of protons as well as alpha-particles of other than radioactive origin appeared, apparently as the result of cosmic-ray disintegrations. Careful inspection of 60 sq. cm of plates covered with other materials, however, failed to reveal any energetic protons or alpha-particles (R6).

Iford New Halftone plates, left for 5 months on the Hafelekar at an elevation of 2300 m by Blau and Wambacher (B19), yielded about 170 proton tracks per sq. cm. Using an emulsion about seventy microns thick, these workers observed some tracks longer than 1 m air equivalent. Although their plates were not covered with paraffin, at least some of the protons they observed are attributable to neutron recoils, for the gelatine in the photographic emulsion is rich in hydrogen.

Further evidence for neutrons was obtained by Schopper (S3), who sent some paraffin-covered Agfa K-plates into the stratosphere in an unmanned balloon. They remained at an average altitude of 17.8 km for 6.5 hours. Comparing the frequency of proton tracks on these plates with that of plates left at 3400 m, Schopper counted 5.1 and 0.28 tracks per sq. cm per hour, respectively, at the two elevations. The frequency near sea level was 0.07 track per sq. cm per hour.

To detect slow neutrons—whose energy is insufficient for the ejection of recoil protons—Schopper applied the well-known boron reaction, in which alpha-particles are emitted. For this purpose he used the Ilford R5 emulsion, impregnated with a boron compound. Measurement of the alpha-tracks found in the emulsion disclosed the presence of a group of alphas whose air-range was about one cm. The plates exposed at 200 m above sea level showed 0.15 alpha per sq. cm per hour as compared with a frequency of 0.25 track at 3400 m.

More recently, Heitler, Powell, and Fertel (H6), have studied the relative absorption in air and lead of the radiation responsible for cosmic-ray protons. Plates covered with 13-cm lead, as well as unshielded plates, were left on the Jungfrau for 230 days; these were compared with controls left at sea level. The unshielded Jungfrau plates had about ten times as many proton tracks as those at sea level, but only slightly more than the lead-covered emulsions. Thus electrons and mesotrons were both ruled out as the primary rays producing the tracks; the former because they would be completely absorbed by the thick lead, the latter because their intensity increases by a factor of only 1.7 between sea level and 3400 m. It was concluded that the protons are ejected by a third component, probably neutrons. (For a general discussion of cosmic-ray neutrons, see B3.)

This view was modified after a more detailed investigation by Heitler, Powell and Heitler (H7). They obtained track-frequency data from emulsions shielded by six different thicknesses of lead on the Jungfrau. Their absorption curve shows a maximum at 1.2 cm, and in general bears a striking resemblance to the Rossi transi-

tion curve.³ It seemed likely, therefore, that while some of the protons found in emulsions are produced by neutrons, there is another group due to the soft component. A plausible mechanism is suggested for this process—a nuclear photoelectric effect, in which a cosmic-ray photon releases a proton from the nucleus.

In the foregoing investigations the tracks of heavy particles were treated as isolated events, and their mode of origin was interpreted in terms of this conception. In the light of other evidence from photographic emulsions, however, it is probable that a significant portion of the “single” tracks really stem from multiple nuclear disintegrations. On plates sent up in an unmanned balloon flight, Wilkins had observed two groups of associated particles, in each of which tracks emerged asymmetrically from a common origin above the emulsion. He considered these to be proton showers.

Many examples of the simultaneous ejection of several particles from a nucleus were obtained by Blau and Wambacher (B20, B21). On their Hafelekar plates they found groups of tracks radiating star-like from centers within the emulsion. These “stars” represent disintegrations far more energetic than those commonly observed in the laboratory. In most artificial nuclear reactions, a single particle is emitted from the activated nucleus; cosmic-ray disintegrations, on the other hand, have yielded as many as fourteen ionizing particles in a single excitation, and any neutrons released in the same event would have left no trail in the photographic emulsion.

Wambacher (W2) made a detailed study of 154 stars, not counting two-particle events. It was observed that most of the tracks are incomplete, i.e., they do not terminate within the layer of gelatine, but above or below it. Therefore, the longer tracks, being more likely to represent complete paths within the emulsion, were used in plotting an empirical curve of grain density against range. The star tracks seemed to be produced by one kind of particle. A comparison with proton tracks of energy up to 10 Mev—ejected from paraffin by beryllium neutrons—showed that the particles are probably protons.

³For a discussion of the Rossi curve, see Froman and Stearns, *Rev. Mod. Phys.* **10**, 143 (1938).

It should be noted that this by no means excludes neutrons as products of the disintegration process, for neutrons have too low a specific ionization to affect a series of silver halide grains.

From the calibration curve and the incomplete star tracks, the range of each particle was determined from the grain density of its track; thence the energy of the particle was deduced. Total disintegration energies, obtained by adding the energies of constituent tracks, were not uncommonly greater than 60 Mev. In the most energetic event observed, the sum of the proton energies was 150 Mev; this figure must be doubled if we make the reasonable assumption that a number of neutrons comparable with the number of visible tracks was emitted.

In a series of unmanned balloon flights, Schopper and Schopper (S4) succeeded in keeping some Agfa K-plates floating high in the stratosphere for several hours. In addition to long proton tracks, they found star tracks whose grain spacing indicated that they were due to alpha-particles. Unlike Wambacher, therefore, they used two calibration curves in their energy determinations—one for protons, the other for alpha-particles. Occasional tracks, moreover, showed so high a grain density that they were attributed to nuclear fragments more massive than alpha-particles. Thus one of their five-fold disintegrations included three alpha-particles, one proton, and a particle described as “at least as heavy as an alpha.”

The presence of alpha-particle tracks in disintegration stars was independently observed in an extensive investigation by Filippov, Jdanov, and Gurevich (F1). Utilizing the technique and the highly sensitive “E3 emulsion” developed by one of them (J1), they counted several thousand single tracks and stars comprising from two to five tracks. They do not give the elevations at which the various data were obtained, but apparently most of the plates were kept at their laboratory in Leningrad. Some of their emulsions were impregnated with boron, and these showed a relatively higher track frequency than the untreated plates.

The observed stars were classified in two categories: (a) disintegrations in which the particles emerge in random directions from a

common center; (b) groups of tracks emanating from a common point, but confined within a relatively small solid angle. The latter events, designated as "proton showers," occur especially often in glass. The Russian workers recognize that these showers may be fundamentally not different from the random type of disintegration. They incline, however, to the view that a special mechanism may be required to explain the proton showers.

Filippov and his associates attempted, from considerations of energy and momentum conservation, to identify the incident ray which gives rise to a star. First they computed, for a given disintegration, the total energy and the vector sum of the constituent momenta. Then they tried in succession various combinations of assumptions regarding the kind of nucleus disintegrated, and the character of the incident particle which had produced it. For example, in the case of a threefold disintegration observed in an emulsion containing borax, the guess was first made that the element disrupted was boron. It was further supposed that the bombarding particle was a neutron. On these assumptions several nuclear reactions were tested in an effort to find one which would meet the requirements of energy and momentum balance. When isotope B^{10} had been ruled out, the B^{11} nucleus was tried, but with no greater success. The bombarding particle was then assumed to be a photon instead of a neutron. It turned out that this guess also failed to meet the requirements. Consequently, the boron hypothesis was rejected, and the same trial-and-error sequence was applied to a heavier nucleus, until a reasonable accord with the experimental data was achieved. The same procedure was tried out for a number of stars. In these calculations, the possibility seems to have been overlooked that, in addition to charged particles, a number of unrecorded neutrons may have been liberated in the disintegration. If this were the case, the energy and momentum sums would, of course, be quite different from the computed values.

It must be evident that this procedure is fraught with uncertainties. In the first place, the emulsion contains a variety of nuclei: silver, bromine, nitrogen, oxygen and carbon. To exhaust the possibilities, one would have to con-

sider all of their isotopes. The identification of the visible tracks is by no means certain. Thus, while one can frequently decide between a proton and an alpha-particle, it is much more difficult to distinguish these from an H^2 , an H^3 , or an He^3 particle—any one of which may, for all we know, occur in a star. Finally, the entire calculation is based upon the assumption of the simultaneous emission of all the particles. That such is often the case seems plausible from theoretical considerations (to be discussed below). But in any given event one can hardly rule out the possibility that a step-wise, rather than a simultaneous disruption has occurred. The Russian workers were aware of the many arbitrary assumptions involved in this method; accordingly, they refrained from drawing any general conclusions as to the nature of the disintegrations.

That the frequency of disintegration processes seems to rise rapidly with increasing altitude had been noticed by several observers. Stetter and Wambacher (S7) investigated the variation with altitude of the number, energy and multiplicity of stars. They studied several hundred stars in plates which had been exposed at six different elevations, from 200 to 3400 meters. The frequency varied most markedly with the altitude. The average total energy per star increased approximately twofold between 1000 m and 2000 m above sea level; while the average number of tracks per star increased slightly. These results indicate that the radiation producing the stars is strongly absorbed in air. It seems natural, therefore, to identify it with the soft component.

While it was admitted that some of the tracks may be due to alpha-particles, a protonic origin was considered more probable; and, for convenience, all the tracks were ascribed to protons. In computing the total energy per star, the Vienna workers assumed that neutrons equal in number and total energy to the protons were emitted in each event. Moreover, the binding energy of each particle was taken into account. Thus the total excitation energy was given by twice the total kinetic energy of the protons plus the total binding energy. On this basis the energy of a typical star turned out to be 60–70 Mev, although an appreciable number of

disintegrations involved as much as several hundred Mev.

Working on the hypothesis that the star-producing radiation consists of particles, and that the various disintegrations are due to a single kind of particle, Stetter and Wambacher tried to deduce the order of magnitude of its mass. They computed the vector sum of the momenta, assuming the magnitude and distribution of the neutron momenta to be similar to those of the protons. In order to assign an appropriate momentum to the residual nucleus, one must have some idea of its mass. Accordingly, a star with seven or more tracks was ascribed to a silver or bromine disintegration, and the average of the masses of these two elements was assigned to the parent nucleus. Similarly, for smaller disintegrations, the average mass of the lighter nuclei in the emulsion—oxygen, nitrogen and carbon—was used.

In terms of the total momentum, P , and the total energy, E , the mass is given by $P^2/2E$. Mass values were obtained for more than thirty stars, and the frequencies of the various masses were plotted on a block diagram. As interpreted by the authors, the results indicated that the most probable mass of the primary particle is closer to the proton mass than to the masses of any of the other elementary charged particles. The evidence, therefore, seemed to point to neutrons as the star-producing radiation. Undoubtedly the method is invalid when applied to single events, but unlike the procedure of the Russian workers, the approach of Stetter and Wambacher is a statistical one. It is not unreasonable to expect a mass value of at least the right order of magnitude.

We consider next the mechanism by means of which a cosmic ray can cause a multiple disintegration. Frenkel (F2) and Bohr (B24, B25, B26) suggested that a process analogous to evaporation may be responsible for the simultaneous emission of several particles. According to Bohr's hypothesis, the initial encounter is followed by the formation of a semi-stable compound nucleus, with a partition of the excess energy among a number of nuclear particles. When these have acquired sufficient kinetic energy to overcome the potential barrier, they escape from the nucleus. Heisenberg (H1a, H2)

concurs in the view that the energy made available by the incident ray raises the temperature of the entire nucleus. The result of this heating is an intense evaporation and radiation. In general, neutrons are most likely to be emitted, as they do not need to overcome the Gamow potential barrier. Alpha-particles have twice as high a Gamow barrier as do protons; hence their emission is less frequent.

For the emission of a given particle, Heisenberg, as well as Bagge (B1), consider two alternative mechanisms: (a) The particle may be "evaporated" after the nucleus has been heated up; (b) if the particle lies near the surface of the nucleus, it may be ejected directly. If a track is due to the latter type of event, we should expect it to show a higher energy than the average, and a preponderantly downward direction. From an analysis of the more energetic tracks in their emulsions, Stetter and Wambacher obtained a random distribution in direction. They concluded that the longer tracks are also produced as a result of evaporation, and not by direct impact.

Heitler (H5) calculates that a 100 Mev excitation of a heavy nucleus results in the emission of about twenty particles. One-half of these or more are neutrons; hence about five to ten are protons, in good agreement with the number of tracks found in stars of high energy. Bagge (B1), basing his treatment on Heisenberg's analysis (H2) and the data of Blau and Wambacher, estimates the range of nuclear forces. He also discusses the relative intensity of the neutron radiation to be expected if equal numbers of neutrons and protons are ejected in cosmic-ray disintegrations. He concludes that because of their greater range, neutrons in the atmosphere should outnumber protons 250 : 1. (Also see G2, H3, H4, B4, U1).

Besides the well-confirmed findings relating to single tracks and stars, several other phenomena observed in plates deserve mention. Filippov, Jdanov, and Gurevich (F1) report some tracks of anomalously large grain spacing associated with their proton showers. They think it is likely that these are mesotron tracks. Bose and Chowdhry (B27) attribute fifteen narrow-angle pairs of tracks found in vertically oriented plates to mesotrons. A burst of one hundred heavy particles was found by Jdanov (J5, J6)

in a plate exposed at a high altitude. That the event was not caused by radioactive contamination is gathered from the fact that some of the tracks in it have an air equivalent range of 18 cm. Taylor, Fraser, and Dabholkar (T10) impregnated a part of the area of some plates with samarium sulphate, and exposed them at an altitude of 8000 ft. A study of certain alpha-tracks—believed to be due to the radioactivity of samarium—in the treated plates led to the remarkable conclusion that the cosmic rays influence the rate of decay in that element. (Other photographic emulsion phenomena attributed to cosmic rays are reported in the literature; see, e.g., H7, J2, J3, J4, T2.)

IV. CONTRIBUTIONS OF THE PHOTOGRAPHIC METHOD TO OTHER PROBLEMS IN NUCLEAR PHYSICS

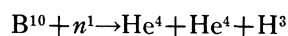
The photographic emulsion technique has been successfully applied to the study of nuclear phenomena other than those of cosmic-ray origin. Most of the heavy products of natural and artificial disintegrations—protons, alpha-particles, deuterons, and even fission fragments—have been registered on plates, while neutrons have been detected by means of the recoil proton tracks they produce.

As mentioned in Section II, proton tracks were first obtained in 1925 by passing polonium alphas through a thin layer of paraffin (B5, B6). The lengths of these recoil tracks were measured by Riedl (R3), who found four range groups of protons. After the neutron was discovered, beryllium neutrons were used to eject more energetic protons from paraffin (K4, B15). Kirsch and Wambacher (K4) observed fifteen velocity groups of beryllium neutrons, distributed almost uniformly from 10^9 to 5×10^9 cm/sec. More recently, Powell and Fertel (P2) have applied the recoil proton method to the study of neutrons released from various light elements bombarded with deuterons.

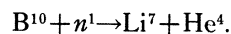
Protons have been detected not only as recoils, but also as the products of disintegrations. Reference has been made above to the work of Blau (B5, B6, B7, B8) on the disruption of aluminum and carbon by polonium alphas. The aluminum reaction in particular has been extensively

studied. Blau and Wambacher (B14, B15, B16) identified groups of protons corresponding to those which Constable and Chadwick had found with a valve amplifier, but also observed an additional group not previously reported. Step-pan (S6) resolved into eight groups the protons emitted from aluminum at right angles to a polonium-alpha beam. Merhaut (M4) measured the proton yield as a function of the alpha-energy, and found four resonance peaks corresponding to alpha-ranges of 3.6, 3.2, 2.8, and 2.4 cm.

Several types of disintegrations produced by neutrons have been studied by the photographic method. For the disintegration of boron by slow neutrons, Taylor and Goldhaber (T6) expected the reaction



to be more likely than



When a plate was soaked in a borax solution and then the dried emulsion exposed to a source of slow neutrons, only single tracks were registered. Since at least some threefold forks would have been produced by the first reaction, it was concluded that the disintegration proceeds according to the second scheme. Later Taylor (T5), using fast neutrons, found that the three-particle reaction does occur, although very rarely compared with the other process. Gurevich (G1) determined the upper energy limit of neutrons emitted spontaneously by the artificially radioactive disintegration products of phosphorus. Burcham and Goldhaber (B28) obtained evidence that the disintegration of N^{14} by slow neutrons yields C^{14} and a proton. They used photographic emulsions of different sensitivity to distinguish between the emission of alphas and protons.

Photographic investigations of nuclear reactions excited by deuterons have been reported by several workers. Powell and Fertel (P2) bombarded boron with deuterons and directed the emitted protons tangentially into a plate. They obtained a range-frequency curve from the measured track lengths, and claimed a higher resolving power for this method than that obtained with the cloud chamber. Richards and Hudspeth

(R2) have studied the photographic plate spectrum of the neutrons liberated when deuterium is bombarded with deuterons.

Most of the nuclear experiments in which the photographic technique has been used have been concerned with natural radioactivity. In 1930 Blau (B9) found that the emulsion tracks of ThC alphas agreed in number and distribution with those obtained with a tube electrometer. Halbwachs (H1) also studied the alpha-spectra of ThC and ThC' photographically. The emission of a series of alphas corresponding to the successive members of a radioactive series may result in a group of tracks radiating from a point—resembling the disintegration “stars” produced by cosmic rays—according to Taylor and Dabholkar (T9) and Wilkins (W7). The latter “infected” emulsions with drops of solutions of radium and thorium active deposit. The track due to RaC' was found always accompanied by another track making a characteristic angle of 110° with it. Since this result disagrees with the predictions of disintegration theory, Blau and Wambacher (B17) repeated the experiment. They were unable to confirm Wilkins' findings, but pointed out that their method of angle-measurement was not as precise as his.

On plates impregnated with samarium Taylor (T7) found, in addition to the short range alphas previously known, a group of particles with air ranges up to 3.5 cm, believed to be protons. He concluded that samarium emits singly charged particles. The apparent influence of cosmic radiation on the alpha-activity of samarium has been mentioned in the last section (T10). Wilkins and Dempster (W10) identified the radioactive isotope of samarium by microscopic examination of a plate exposed in Dempster's mass spectrograph and stored for many months. Wilkins, Rayton and St. Helens (W4) measured alpha-particle speeds by impressing a high frequency on two parallel rods and passing the rays longitudinally between them. Ringo (R3a) built a magnetic spectrograph capable of measuring alpha-particles of energies from 2 to 20 Mev.

Using plates to detect the particles, he measured the energy of the main group of protoactinium alphas. In all the alpha-spectra he investigated, large numbers of particles with energies below those of the main groups were found.

A scattering camera, in which plates are arranged fanwise, was developed by Wilkins and Kuerti (W12) for the study of nuclear scattering. Powell, May, Chadwick, and Pickavance (P3), using another kind of “camera,” investigated the scattering of protons by light, gaseous elements. For the Ne nucleus, an excited state of 1.4 Mev, attributed to inelastic scattering, was observed.

The photographic detection of nuclear fission was accomplished by Myssowsky and Jdanov (M9), who exposed a plate above a coat of metallic uranium powder to a cyclotron beam of neutrons. In addition to the alpha-tracks due to the natural decay of uranium, they found some short tracks, thicker and more continuous than the former, which they ascribed to the recoil nuclei produced by the fission of uranium. No recoil nuclei were observed after neutron irradiation of bismuth, gold, and platinum for three hours.

The value of the photographic plate as a labor-saving device has been aptly described by Powell and Fertel (P2):

For the analysis of . . . neutrons from the Li+D reaction by the expansion chamber method, 20,000 stereoscopic pairs of photographs were taken. Of those, 1600 tracks were suitable for measurement. We have measured the energy distribution of these neutrons by the photographic method. The 3000 tracks for the analysis are all contained in 3 sq. cm of an Ilford halftone plate obtained in a single exposure of a few minutes.

It is no wonder that Powell and Fertel conclude, “. . . the results . . . suggest that the method merits the serious consideration of those engaged in nuclear research.”

In conclusion, the author takes pleasure in expressing his thanks to Professors Arthur H. Compton and George S. Monk for their valuable guidance and encouragement.

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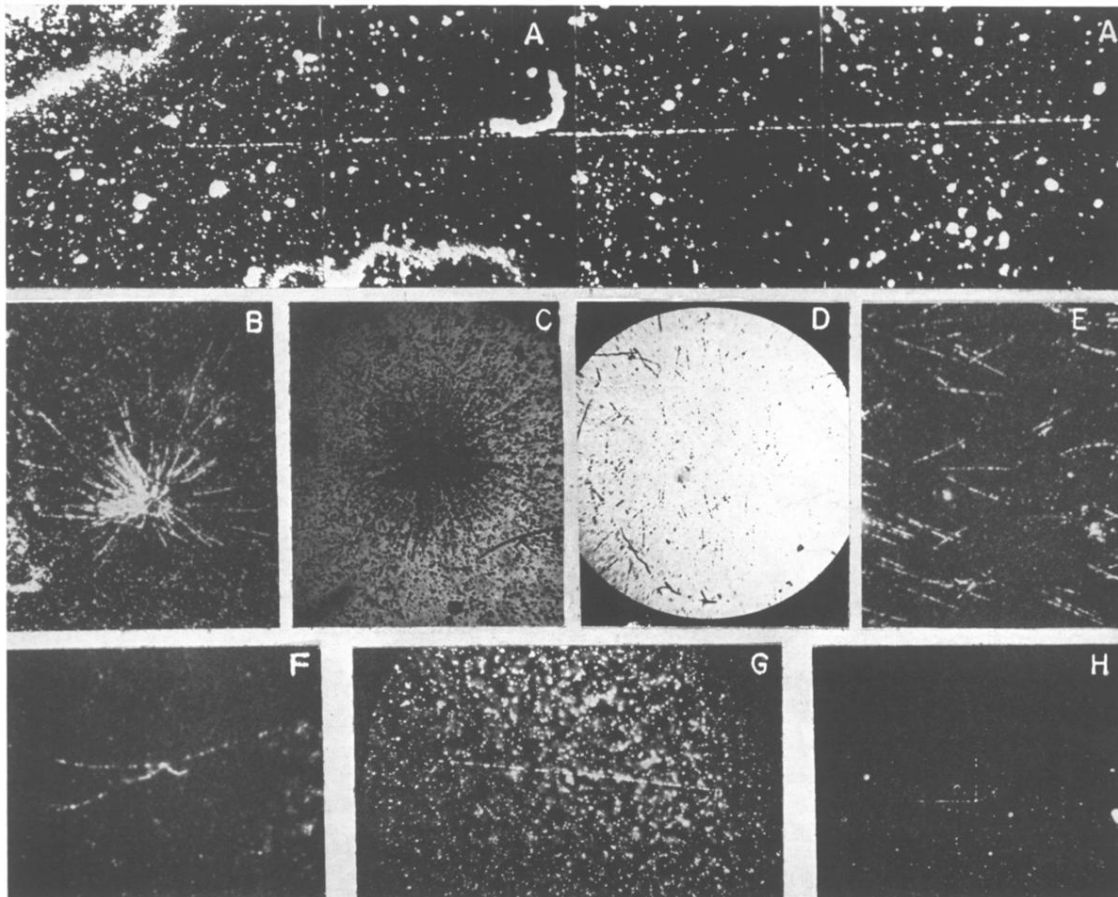


FIG. 1. (A) Cosmic-ray track from plate sent up in stratosphere balloon flight by T. R. Wilkins. Length of track in photographic emulsion, 0.55 mm. Interpreted from the grain spacing as a very high energy alpha-particle, whose range in air is about 75 cm. Magnification 260X.

(B) and (C) Alpha-rays emanating from "contamination centers" in an emulsion impregnated with polonium. Black streak in (C) is a fiducial mark in the microscope eyepiece.

(D) and (E) Po- α tracks obtained by exposing plate edgewise to a radioactive source. Thick black marks in (D) are caused by dust particles on the plate surface. A comparison of photographs (B) and (E) with (C) and (D) gives some indication of the advantage of dark-field illumination over bright-field. Of the latter pair, (C) is from an ordinary process plate; it shows a high background of fog; (D) was obtained with an Eastman Fine Grain Alpha-Particle plate.

(F) Fivefold fork due to successive disintegrations of a single radioactive atom or to alpha-particle emission from several contiguous atoms. At the lower left a short track is superposed over a longer one.

(G) Cosmic-ray track of an alpha-particle from plate exposed at an elevation of 6700 feet. Length in emulsion, 157 microns; range in air, about 22 cm.

(H) Threefold nuclear disintegration probably due to cosmic ray, from Ilford plate sent aloft into the stratosphere. To render the three tracks simultaneously visible although they do not lie in same plane, low magnification is used. The vertical track appears to be due to a proton; the horizontal one is an alpha-track. (Photographs (B), (E), and (F) are reproduced through the courtesy of Dr. Niel F. Beardsley.)