Neutron Emission by Excited RaD Nuclei

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Neutron emission from an active deposit of radium emanation on pure heavy metals was investigated. It is shown that the observed neutron activity is related to disintegration of the RaC″ isotope. The neutrons are apparently emitted by excited RaD nuclei produced as a result of β disintegration of RaC″. Control experiments have been carried out which permit one to estimate the contribution of other possible processes to the neutron radiation from the shortlived active radon deposit.

POLLARD[1] and Copeland and Lind[2] have mentioned the existence of a weak neutron background for sources prepared from radium, radiothorium, or radon. This background has been attributed to the interaction of α radiation, emitted by radium, radiothorium, or by their disintegration products, with the backing material or to the interaction of α radiation with the medium surrounding the source. Recently[3,4] it has been shown that the neutron radiation is mainly connected with the disintegration of the RaC″ isotope. It was shown that it is energetically possible for neutrons to be emitted by excited RaD nuclei produced as a result of the β disintegration of RaC″. Neutron activity with a corresponding life time has been observed.

1. EXPERIMENTAL METHOD

a) *Neutron Measurements*

This paper is devoted to an investigation of the neutron emission of preparations of RaC″, where one has about 3 neutrons per 1 millicurie/sec of radon, and also, to controlled measurements which permit an estimate of the possible contribution of the other processes to the neutron emission from the shortlived radium deposit.

A large volume ionization chamber, filled with boron trifluoride (Fig. 1a), and a set of proportional counters (Fig. 1 b) were used to measure the number

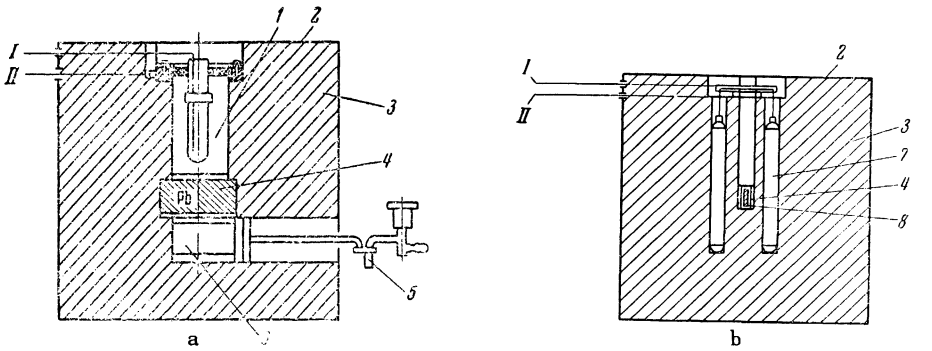

FIG. 1. Experimental arrangement for measuring neutron activity. I — lead to the linear amplifier, II — lead to the high voltage source; 1 — ionization chamber, 2 — screen, 3 — paraffin block, 4 — lead filter, 5 — trap, 6 — chamber with source, 7 — neutron counter, 8 — ampoule with source.

of neutrons emitted by the sources under study. The chamber and source were placed in a large, carefully screened paraffin block; the γ activity of the sources amounted to as much as 400 millicuries. To reduce the γ background a lead disk 80 mm thick was placed between the chamber and the source. During maximum source activity the chamber did not register counts from fluctuations in the γ background.

When the proportional counters were used for measurements, the sources had a γ activity of 1 millicurie or less and were situated at the center of a group of six counters. The diameter of the counter body was 30 mm, the diameter of the central filament was 56 μ, and the length of the operational volume was 250 mm. The counters were operated at a voltage of 1800 v with a gas amplification of about 50; under

these conditions, the γ ray pulses were not registered. The γ activity was measured by means of comparison with a 29.3 millicurie radium standard.

The neutrons were measured by comparison with (Ra-α-Be) neutron standards with activities from 0.5 to 1.5 millicuries (of radium). In the described apparatus the probability of neutrons being registered by the detector depends on their energy and the alignment of the detector and source in the paraffin block. The group of counters used had a stable intrinsic background of about three counts per minute and permitted the detection of radiation from constant sources emitting 2-3 neutrons per second.

Since the probabilities of registering neutrons of different energies emitted by the source and the standard are unknown, the obtained values of specific activity contain a systematic error, which could not be determined because of the low activity of the sources. With comparative measurements on sources of the same type, under standard conditions, this error is eliminated. Non-calculable errors enter in the comparison of data obtained under different geometric conditions or for sources with different neutron energies. For this reason, only the statistical errors of measurement will be shown henceforth.

b) *Preparation of the Radioactive Sources*

The neutron emission of ampoules containing radon and the neutron activity of a short-lived deposit of radium or of preparations of RaC enriched with the RaC″ isotope have been measured. In the first case the radon was inserted into lead ampoules with pressed tubes of various metals. In some cases the ampoules used had nickel seals, which made it possible to inject or withdraw the radon under pressure (Fig. 2).

The radon, which came in glass ampoules, was distilled into a system for further purification, and then was frozen with liquid air so that it could be inserted into a lead ampoule which had been evacuated and outgassed. The short-lived deposit was obtained by using an electric field to activate the foil of the investigated metal in a radon atmosphere. The foil, activated on one side, was placed near the center of the cylindrical vacuum chamber, whose walls were covered with the same foil material (Fig. 3). Measurements of the neutron activity of the short-lived deposit usually took 1.5-2 hours.

The RaC″ preparations were obtained by collecting recoil ions. The active deposit, precipitated onto nickel disks, was allowed to age for about 30 minutes so that the RaA isotope, which emits α ($T_{1/2} \sim 3$ min), would have essentially disappeared. After this time the deposit would contain mainly the isotopes RaB, RaC, RaC′ and RaC″.

Removable nickel plates 0.1 mm thick were placed opposite the disks. A positive voltage was applied to the disk with a field gradient as high as 6–7 *kv*/cm; this served to extract primarily the RaC″ ions produced by the α disintegration of RaC and in addition ions of the long-lived RaD isotope ($T_{1/2}$ = 22 yrs.) produced in the α disintegration of RaC′ (see Fig. 4). Besides these ions, RaC recoil ions

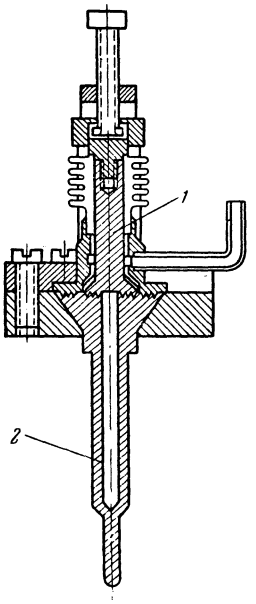

Fig. 2

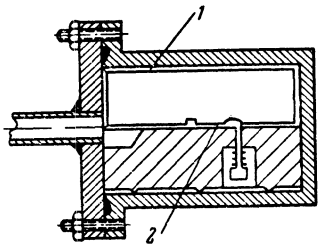

Fig. 3

FIG. 2. Lead ampoule for measuring the neutron activity of radon sources. 1—nickel, 2—insert, Pt foil

FIG. 3. Chamber for measuring the neutron activity of preparations of Ra(B +C +C′) in helium under pressure. 1—outer facing, Pt, 2—specimen

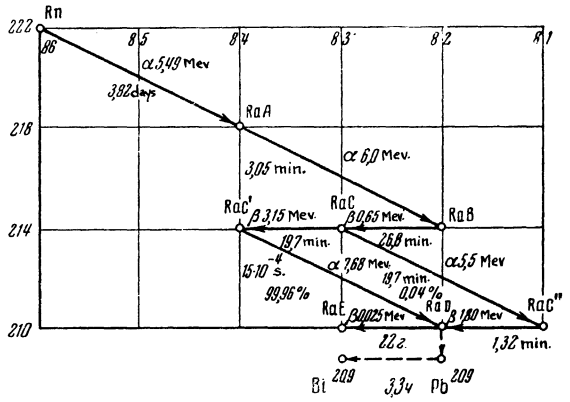

FIG. 4. Radioactive decay scheme for radon disintegration

produced in the β disintegration of RaB also strike the plate. However, the probability of the emission of ionized RaC recoil atoms from the surface of the source is comparatively small, because the recoil energy in a β disintegration is on the order of three times less than in an α disintegration, in spite of the fact that the number of RaB disintegrations exceeds that of RaC α disintegrations by more than three times. The activity of a RaC" preparation increases with larger extracting fields, but at the same time a noticeable quantity of RaC (up to 0.5 millicuries) is collected on the plates.

Each nickel plate was activated for 4.5 minutes, after which its neutron activity was measured. During these measurements the next plate was activated, and so on.

During the measurements of neutron emission the plates were enclosed in nickel envelopes and placed in a small glass vessel which was pumped out to a pressure of 10^{-1} mm of mercury in order to prevent any (α, n) reaction by RaC' α particles with the nitrogen and oxygen of the air.

2. DETERMINATION OF THE NEUTRON EMITTING ISOTOPE

In order to identify the isotope connected with the observed neutron emission, it was expedient to measure the neutron emission of the separate isotopes produced during radon disintegration. Sorting these by chemical methods requires considerable time (which lessens the activity of the preparations) and may lead to the appearance of contaminations by light elements, *i. e.*, an extraneous neutron background.

The change in neutron emission as a function of time was investigated with radon inserted in a clean ampoule and with it pumped out of the ampoule. A comparison of the computed curves for the time dependence of the concentration of different isotopes with the experimental data (Fig. 5) shows that the observed effect is due to RaC or to the short-lived products of its disintegration, RaC' and RaC". The divergence in the computed and experimental data for the neutron activity and α emission at the times when radon was inserted and pumped out is explained by the fact that an active radon deposit is partially carried off.

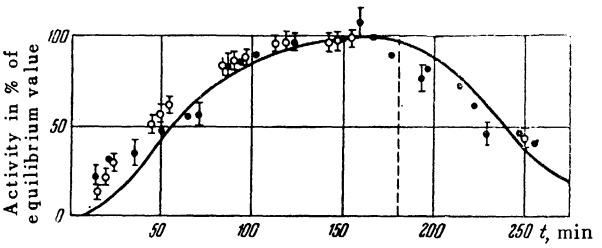

FIG. 5. Dependence of neutron and γ activity of radon on time. $t=0$ — clean ampoule filled with radon; $t=180$ min. — radon pumped out. o — Rn in glass tap (for γ rays). o — purified Rn in a Pt ampoule (for neutrons); solid curve as computed for RaC.

Neutron emission from an active radon deposit may be connected with the following processes: 1) an (α, n) reaction due to RaC' α particles; 2) a (α, n) reaction due to α rays emitted after the β transitions in RaC"; 3) spontaneous fission of the nuclei of one of the isotopes RaC, RaC', and RaC", or RaD; 4) neutron emission by excited RaD nuclei formed during the β disintegration of the RaC" ("delayed neutrons").

Control experiments, described in Sec. 3, showed that the first three processes cannot cause the major portion of the neutrons observed in the experiment. Thefore, there was good reason to measure the neutron activity of an isolated RaC" preparation. When "delayed" neutrons are emitted, the activity should decrease with the half-life period of the β activity of the initial nucleus (the RaC" isotope), *i. e.*, 1.33 min.

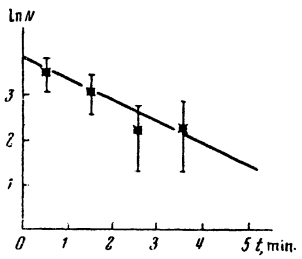

FIG. 6. Time dependence of the neutron actibity of preparations enriched with RaC"

It was possible to perform a series of measurements on 6 to 10 samples enriched with RaC″ before the activity of the deposit had decreased to 50–30% of the initial activity. Figure 6 shows the data for one of the series of measurements. It should be remarked that after the first minute the measured signal exceeded the intrinsic background of the apparatus by less than 4 times. Therefore, the obtained date were summarized for all the samples, were processed for the series as a whole, and the results were plotted on a semi-logarithmic scale. The half life was determined by the method of least squares. The experimental errors were calculated with allowance for the background of the apparatus according to the formula proposed by Peierls[5].

The table below brings together the neutron activity data for measurements on 13 series of RaC″ preparations. Column 5 shows the coefficient of the enrichment of the preparations with the RaC″ isotope in comparison with its equilibrium content in the active radon deposit. The quantity of RaC″ was determined from its neutron activity. In this connection it was assumed that the RaC″ contained in 1 millicurie of radon would emit ~ 3 neutrons per second and that the efficiency of the detector in these measurements (whose geometric conditions varied from the standard) would not differ from the usual efficiency. The activity of the RaC was determined from its γ emission after disintegration of the RaC″. The sixth column shows the "isolation coefficient"—the ratio of the average RaC″ activity in the samples to its activity in the original active deposit. The isolation coefficient, as well as the enrichment coefficient, was determined to within a certain systematic error due to inaccuracy in measuring the number of neutrons emitted by the preparations. From the tabular data it is apparent that, within the limits of the (statistical) measurement errors, the average weighted value for the half life of neutron activity coincides with the half life of RaC″ β activity. This coincidence confirms the assumption that the observed neutron activity is connected with the disintegration of the RaC″.

In order to eliminate the possibility of instrumental effects, control experiments were carried out.

a) The sign of the electric field that extracted the ions was altered in such a way that negatively charged particles were collected on the nickel plates. This done, the neutron activity of the plates fell below the sensitivity range of the apparatus.

b) To check the operation of the detecting system a series of measurements were made using the activity of the *(Po-α-Be)* source, which emitted ~ 10 neutrons per second. These measurements were made under exactly the same conditions as prevailed when the RaC″ preparations were measured. The magnitude of the measured activity of this source did not depend on time.

The data given above and the control experiments show that neutrons are emitted when RaC″ disintegrates.

Neutron Activity of preparations enriched with RaC″

Series Number	Ratio of Signal to Background During First Minute	Number of Samples in Series	Half-Life in min. $T_{1/2}$	RaC″ Enrichment Coefficient of Samples	Isolation Coefficient in %	Avg. Activity of Rac in Samples in Millicuries
1	1	8	1,4 ± 0,7	100	10	0,1
2	0,6	6	0,5 ± 1,9	50	5	0,5
3	0,8	10	1,1 ± 1,9	50	8	0,1
4	0,6	12	3,3 ± 1,9	150	5	0,03
5	0,3	5	2,6 ± 2,9	100	2	—
6	1,5	3	0,85 ± 0,4	—	3	—
7	1	7	1,7 ± 0,8	—	5	—
8	1	2	1,7 ± 1,1	—	7	—
9	0,5	4	0,7 ± 0,6	100	3	0,03
10	2	4	1,6 ± 0,6	1000	3	0,003
11	4	7	1,6 ± 0,4	100	15	0,01
12	3	4	1,15 ± 0,45	—	15	—
13	1	6	1,15 ± 0,95	300	15	0,01
Weighted average			1,50 ± 0,25			

3. CONTROL STUDIES

To clarify the role of other possible processes in the observed neutron activity a series of control experiments were conducted.

a) *The possibility of fission by excited nuclei in the Rn deposit.* An evaluation of the life time for the fission process, according to available empirical formulas and curves, indicates that this is hardly probable (the half-life periods are more than 10^{24} years). Nevertheless, the ionization chamber was used to carry out an experimental check* into the possibility of nuclear fission in the active deposit.

It was shown that the contribution of the fission process amounts to no more than 1% of the observed neutron activity, assuming that in the disintegration no more than three neutrons were released.

b) *The (γ, n) reaction.* The probability of this reaction for deuterium in paraffin may be estimated on the basis of the known energies of γ rays from RaC and RaC"; it amounts to $\sim 2.5\ 10^{-2}$ neutrons per 1 millicurie/sec. This estimate was checked experimentally. Around the radon source there was placed an additional lead absorber, which reduced the number of RaC and RaC" γ rays by 75%. Neutron activity decreased by 1.0%; with a statistical accuracy of 4.6% in the measurements. Obviously, γ radiation in this case could cause no more than 8% of the total neutron activity.

In order to provide a more exact determination of the role of γ radiation, experiments were conducted with a preparation enriched with RaC". Measurements were made alternately under ordinary circumstances and with the source surrounded by a layer of heavy water 2 mm thick. In this case the amount of deuterium that could participate in the reaction, in comparison with that contained in the paraffin, was increased by more than 100 times. The observed neutron activity approximately doubled, with an experimental error of $\pm 30\%$. From this it follows that no more than $1-1.5\%$ of the neutrons emitted by the RaC" preparation were due to the (γ, n) reaction.

c) *An (α, n) reaction due to RaC' α particle.* An estimate of the probability of α particles penetration through the Coulomb barrier shows that for metals with $Z > 72$ (Ta, Pt, Pb), which are ordinarily used for preparing sources, the neutron yield must be less than 10^{-9} per 1 millicurie/sec. The actual neutron yield may increase over that calculated on account of the presence of light impurities, but their content would have to be considerable—greater than the total amount of impurities in the pure platinum used for preparing the sources.

The content of impurities in the platinum which would be necessary to explain the observed neutron yield on the basis of an (α, n) reaction is characterized by the following data:

Impurity	Li	Be	B	C	N	F	Na	Mg	Al	Cl	Fe
Amt. in % (by weight)	0,03	0,01	0,1	2	1	0,05	0,1	0,5	0,3	2	3,5

It should be remarked that filling the ampoules with spectrally pure porous platinum did not change the magnitude of the neutron yield. Nickel was also used in the preparation of the sources; the (α, n) reaction is energetically impossible for most of the isotopes of nickel. The neutron yield in this case should not exceed 0.1–0.2 neutrons per 1 millicurie/sec. However, the averaged yield values for the most thoroughly investigated elements (Ni, Ta, Pt) do not differ, within a range of statistical error from 5 to 15%, which is apparent from the following table:

Backing material	Pt	Ta	Ni
Number of neutrons (per 1 millicurie/sec)	$2,75\pm0,15$	$2,95\pm0,15$	$2,8\pm0,4$

*Together with A. N. Baryshev.

The probability is small that the neutron yield for metals with such a great difference in their atomic numbers (from 28 to 78) would be constant if this yield was basically determined by an (α, n) reaction in the basic isotopes or chemical impurities. Gaseous helium was used for a more direct experimental check as to the role of the (α, n) reaction. About half of the α particles emitted by the source impinge on the metal backing bearing the sample, but the remaining 50% of the α particles may be stopped, either totally or partially in helium (in which the (α, n) reaction is energetically impossible), so it is impossible for this portion of the α particles to give rise to neutrons in any material. The range of RaC' α particles in helium is $\sim$ 35 cm, and therefore the gas used was under a pressure of 6 atm. The air content of the helium did not exceed 0.01% by particle density, *i. e.*, its partial pressure was less than 0.5 mm of mercury. The source was located in the central portion of a cylindrical chamber (Fig. 3) 60 mm in diameter. Platinum was used as the material for the backing of the source and the outer facing of the chamber. The control experiments showed that at this pressure the emission of no more than 0.1 neutron per 1 millicurie/sec could be due to an (α, n) reaction in the residual nitrogen. The energy of the RaC' α particles falling on the outer facing was reduced by collisions in the helium to $\sim$ 5.4 Mev. The data displayed in Fig. 5 show that for α particles from Rn and RaA (E_α = 5.5 and 6 mev), the (α, n) reaction is practically absent. Thus neutron emission would have been reduced by about 2 times if it were due to the (α, n) reaction.

In the experiment the neutron yield was 2.5 ± 0.4 when the helium was absent, but when it was present, the yield was 2.7 ± 0.5 neutrons per 1 millicurie/sec instead of a yield of 1.4 ± 0.2 neutrons which might have been expected from an (α, n) reaction. Allowing for the experimental errors, one can consider that the contribution of the (α, n) reaction must amount to less than 50% of the observed effect. The same results were obtained at lower helium pressures.

The experiments with helium are conclusive in those cases where the reaction occurs either in the backing of the source or the facing of the chamber. If it occurred only on the surface of the source (in some impurity appearing during activization), the folding of the inner layer of the source upon itself would increase the neutron yield by about two times, since the layer can only be very thin, and the yield would show a linear increase with the width. In a test the activity of the sample did not vary within the limits of the statistical accuracy of measurement, $\pm$ 10%.

Another control experiment, which made it possible to evaluate the probability of the (α, n) reaction, was the measurement of the neutron background of Th(B + C + C') sources on platinum. The disintegration pattern of ThB and its radioactive products eliminates the possibility of the formation of nuclei with sufficient excitation energy to emit neutrons. When ThC'') disintegrates, hard γ rays with an energy of 2.62 Mev are emitted, and these cause a (γ, n) reaction on the deuterium in paraffin. The energy of the basic group of ThC' α particles is $\sim$ 8.8 Mev. If the neutron emission from radon sources is due to the (α, n) reaction, then it should increase considerably in the change from RaC' to ThC', because the cross section of the (α, n) reaction near the threshold grows exponentially with the energy of the α particles. Measurements have shown that neutron radiation from a Th(B+C+C') source on platinum is equal to 0.3 ± 1.8, *i. e.*, fewer than 2 neutrons per 1 millicurie, which is less than for Ra(B+C+C'). A background of this order might be expected on account of the (γ, n) reaction in paraffin. The small degree of accuracy in the measurements is explained by the low activity of the radiothorium preparation used in the control experiments.

All of the described results show that neither the (α, n) reaction, (γ, n) reaction nor nuclear fission in the active deposit is the basic process responsible for neutron emission from radon sources. The possible total contribution of these reactions to the total neutron emission is difficult to estimate quantitatively because of the low absolute activity of the sources and the short life time of the active deposit.

CONCLUSIONS

The investigations carried out confirm the predictions[3] that the basic process responsible for the neutron activity of a short-lived active radon deposit will be neutron emission by excited RaD nuclei produced during the β disintegration of RaC''. It should be noted that this process is energetically possible. According to the data available on the thresholds of *(d, p), (γ, n),* and *(d, T)* reactions, the binding energy of the neutron in RaD is 5.2 or 4.8 Mev[6]. The low value for the binding energy in RaD is explained by the fact that in RaD there are only 2 neutrons outside the filled shell (*i. e.* Pb^{208}). In the Pb^{209} nucleus one has an even smaller binding energy—3.9 Mev. The total energy of the RaC'' – RaD transition has not been directly measured because it has not been possible to isolate a sufficient quantity of this comparably short-lived isotope. The transition energy can be determined from the known data on the α disintegration of RaC and RaC' and the β disintegration of RaC by comparing the total energy of the conversion of RaC to RaD over two branches of the

"radium fork" (see Fig. 4). Judging by the latest published decay scheme for RaC[7], the total energy of the RaC" – RaD transition is 5.4 Mev. It is obvious that the sum of the energy of the partial β spectrum and the kinetic energy of a neutron is 0.2 or 0.6 Mev, if the magnitude of the binding energy is respectively 5.2 or 4.8 Mev.

According to our data, the probability of neutron emission due to the β disintegration of RaC" is $\sim 2\times10^{-2}$ %. A partial β spectrum is also predicted with an end-point energy of $\sim$ 100 kev and a relative transition probability of $10^{-1} - 10^{-2}$ %. When the energy of the basic β transition is $\sim$ 2 Mev, the existence of this transition is possible. The neutron kinetic energy in this case should be no more than 0.5 Mev.

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Elastic and Quasi-elastic Scattering of 660 Mev Protons by Deuterons *

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The differential elastic p–d scattering cross sections at angles of 40° to 150° and the differential quasi-elastic p–p scattering cross sections at angles of 50 to 90° of the two nuclei were measured in the center of mass system by the ganged-telescope method. The experimental data point to the existence of a predominant interaction between the incident proton and the separate nucleons in the deuteron and also to the existence of collective interactions between the three nucleons. The energy dependence of the differential cross section of quasi-elastic p–n scattering into an angle of 90° in the c. m. s. of the two nucleons was also measured in the 460 to 660 Mev range.

1. A study of the interaction of fast protons with the simplest nucleus—the deuteron—leads to several conclusions concerning the elementary nucleon-nucleon interactions and concerning the character of the motion of the nucleons in the deuteron, and also permits an approach to the study of the collective interaction of nucleons at high energies.

This article describes the experimental results of p–d elastic and of p–p and p–n quasi-elastic scattering of 657±2 Mev protons by deuterons. The proton beam was generated in the synchrocyclotron of the Institute for Nuclear Problems of the Academy of Sciences of the U.S.S.R. The method of ganged telescopes [1] was used to separate the above processes from the variety of meson-producing proton–deuteron interactions. The twice-collimated proton beam from the accelerator passed through a monitor, before striking the target. An ionization chamber filled with helium was used as a monitor in the

*This work was reported at the All-Union Conference on the Physics of High Energy Particles, May 1956 and at the Geneva Conference in June, 1956.