GIANT LIQUID SCINTILLATION DETECTORS AND THEIR APPLICATIONS*

FREDERICK REINES

Los Alamos Scientific Laboratory, Los Alamos, New Mexico

I. GENERAL CONSIDERATIONS LEADING TO THE DEVELOPMENT OF LARGE DETECTORS

WHEN Clyde Cowan and I started in 1951 to pursue the free neutrino,[1] we knew that an essential ingredient in any successful scheme would be a solid or liquid target consisting largely of protons and measuring approximately a cubic meter. Furthermore, the events which occurred in this target had to be viewed in some detail and in an efficient manner, a requirement which suggested a transparent medium. We had heard a little about liquid scintillators, but 'large' was at that time a term applied to an object with a volume of 0.02 m^3.† Early in 1952 we designed and actually built an object which we named 'El Monstro'. Figure 1 is a sketch of this detector which was in the

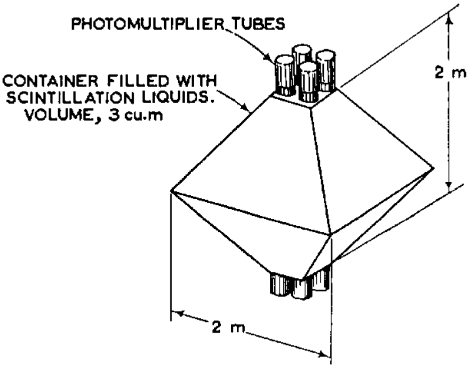

Fig. 1. Sketch of 'El Monstro', first Los Alamos attempt at a giant liquid scintillation detector (1952).

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* Work performed under the auspices of the U.S. Atomic Energy Commission.

† Gamma detectors of this size have been employed by the Los Alamos group of D. W. Mueller since mid 1950. M. R. Cleland and H. W. Koch at the National Bureau of Standards built a slightly larger detector (0.03 m^3) for total gamma-ray absorption studies (Nucleonics **10**, No. 3 (1952)). Their detector was a cylinder 1.2 m long and 0.18 m in diameter which was viewed by 16, 2 in. photomultiplier tubes placed around the cylinder walls. The gamma-rays to be absorbed were directed down the axis of the cylinder. Because of their detector geometry the scintillation light paths to the photomultiplier tubes were short so that absorption by the liquid presented no problem.

shape of two pyramids placed base to base. It was 3 m^3 in volume and had provisions for several 2 in. photomultipliers at each apex. We were later fortunate in being joined by an able group among whom were Anderson, Harrison and Hayes.

A little will now be said about the physics of neutrino detection. The interaction which we used was

$$\nu_- + p \longrightarrow \beta^+ + n \tag{1}$$

where the antineutrino (ν_-) transmuted the proton (p) and made a positron (β^+) and neutron (n). The idea is to have a detection system with many protons to serve as antineutrino targets, and in this case these were supplied by the liquid scintillator. We also required high neutron efficiency and, although at the beginning we didn't think of utilizing delayed coincidences to discriminate against backgrounds, we thought it would be nice if we could observe the positron as well. High neutron efficiency implies not only containing the neutron in the system, but also detecting the gamma-ray resulting from neutron capture.[2] The point here is that the neutrons resulting from reaction (1) are all low energy, $\lesssim$10 keV, so that they could not be seen against background via the recoil protons. We therefore required high gamma-ray efficiency. It happens that a system large enough to absorb capture gamma-rays through the Compton collisions which predominate in the light elements in the few MeV region, has enough target protons to insure several antineurtrino induced events per hour.*

An additional problem arose because of the size of the system. In El Monstro, for example, the scintillation light had a longest distance of 2 m to travel before reaching the detector wall. Since no one had previously asked for scintillation light to travel as much as one meter in the scintillator prior to collection by a photomultiplier tube, it was not clear that this requirement was going to be feasible. At the time (1952), toluene, terphenyl, and the Hayes wavelength shifter, α-NPO, were materials which could readily be made available in quantity and it turned out that this combination using reagent grade toluene was sufficiently transparent. The α-NPO was required to change the wavelength of the scintillation light so that it could travel more readily through the liquid to the liquid boundary. An additional point had to do with matching the scintillation light to the photomultiplier response. Since we wanted to collect a reasonable fraction of this light, we needed a large number of photomultipliers surrounding the liquid. The number of photomultipliers was reduced to manageable proportions by the use of a white (reflective) coating on the inside of the detector tank.

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* The expected rate of events, R, can be calculated from the cross section, $\bar{\sigma}$, for reaction (1) averaged over the antineutrino spectrum for the fission reaction source, $\bar{\sigma} \sim 5 \times 10^{-44}$ cm^2, the antineutrino flux, f, available, f $\sim 10^{13}$ ν_-/cm^2 sec, and the number of target protons, N. $R = \bar{\sigma} \mathrm{f} N$. The rate of detection, Q is the rate of occurrence times ϵ the detection efficiency of the system $Q = \bar{\sigma} \mathrm{f} N \epsilon$.

This, then, is a sketch indicating how the requirements arose for our very large liquid scintillation detectors: they arose from a consideration of what was necessary in order to detect the free neutrino.[3]

Figure 2 is a picture of the second large volume detector designed for neutrino work and used at Hanford in 1953. It is a right circular cylinder 75 cm high, 75 cm in diameter with ninety 2 in. photomultiplier tubes on the cylindrical wall. Five-inch tubes were not known at that time (1952) and 2 in. tubes were just going into mass production. We actually built two of these detectors. In order to minimize tube noise, which is something that worried people a great deal in those days, we divided the ninety tubes into two inter-leaved banks and ran them in coincidence, requiring both banks to see the scintillation light. Despite the coincidence arrangement the tubes had to be carefully selected against double pulsing and noise, and the gains had to be more or less uniform so that a photon striking any one of the tubes would cause the emission of an electron with equal probability. Figure 3 shows the ten compartment tube tester which we used in our most recent work for selecting and balancing photomultipliers. A comment about the reflective coating; the detector walls were made and kept white with great difficulty because of the action of the scintillation mixture. White Tygon remained attached for a time measured only in weeks, a magnesium silicate paste was somewhat more successful. Today we use a titanium dioxide pigmented paint called 'Plasite'.* The point here is that toluene is a mean stuff which was made even more difficult to handle when we incorporated into it cadmium propionate so as to obtain more energy on neutron capture than that available from hydrogen.

Following the Hanford work Cowan and I designed a new experiment which was to employ a much improved and somewhat larger detector. Another group among whom were A. Brousseau, F. B. Harrison, H. W. Kruse, A. R. Ronzio and later A. D. McGuire, was formed for the purpose of doing the experiment at the Savannah River Plant of the Atomic Energy Commission (1956). In the antineutrino identification experiments[1, 4] we observed the antineutrino reaction (1) occurring in a water target 7.5 cm deep and 2 × 1.5 m in lateral extent. Cadmium dissolved in the water in the form of $\mathrm{Cd\,Cl_2}$ served to capture the moderated product neutrons giving rise to several gammas with a total energy of 9 MeV. The β^+ annihilation gamma-rays and the cadmium capture gammas were viewed in delayed coincidence by two giant liquid scintillation detectors placed above and below the water target and with a sensitive volume measuring 60 cm in depth by 2 × 1.5 m in lateral extent. The liquid we used was purified triethylbenzene with terphenyl (3 g/l.) and POPOP (0.2 g/l.). We actually used two target tanks and three such giant detectors having TEB capacity of 470 gal each. Figure 4 shows the 3600 gal capacity tank farm built to contain the necessary liquids plus spare fillings.

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* Obtained from the Wisconsin Protective Coating Company, Green Bay, Wisconsin.

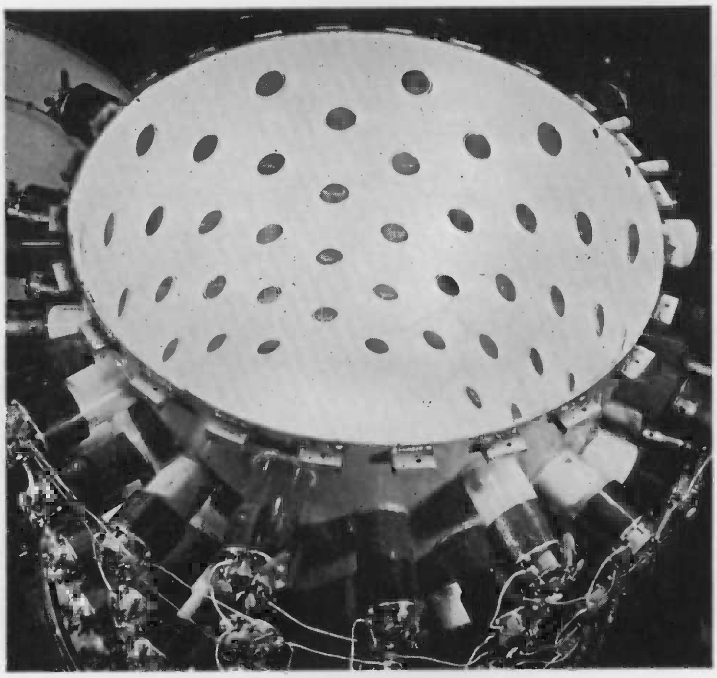

Fig. 2. The Hanford neutrino detector (1953).

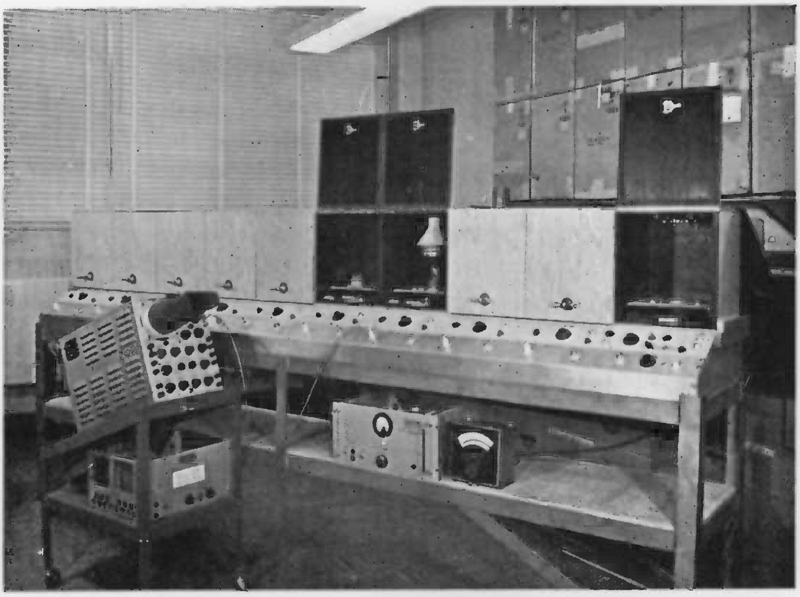

Fig. 3. Gang photomultiplier tube tester for multitube scintillation detectors.

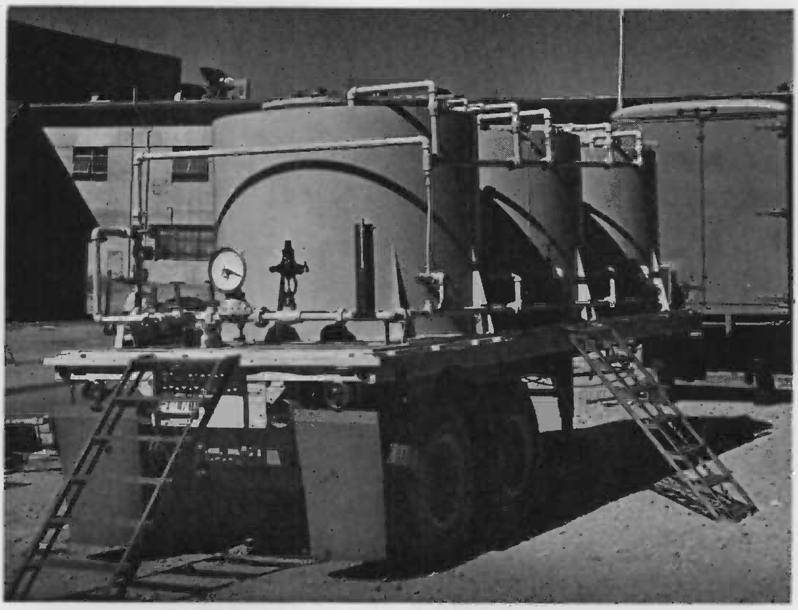

Fig. 4. Portable scintillation liquid storage tanks, 3600 gal capacity.

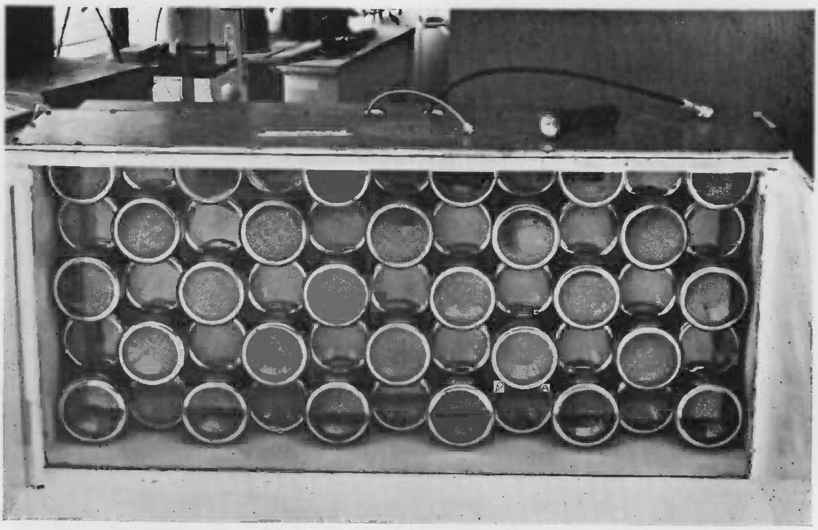

Fig. 5. Inside view of giant detector tank used in neutrino experiments at the Savannah River Plant (1956).

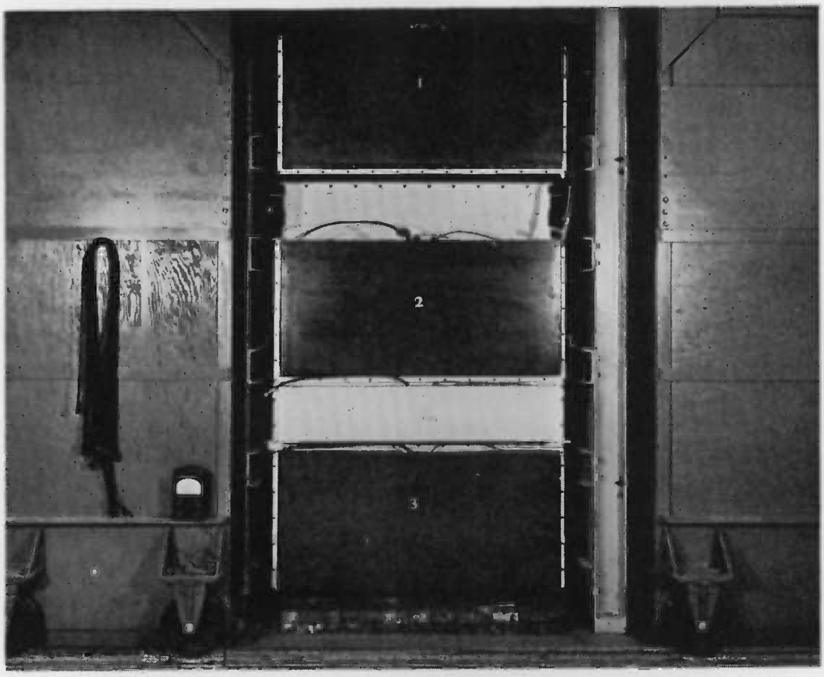

Fig. 6. Neutrino detector array at Los Alamos inside massive lead shield (1955).

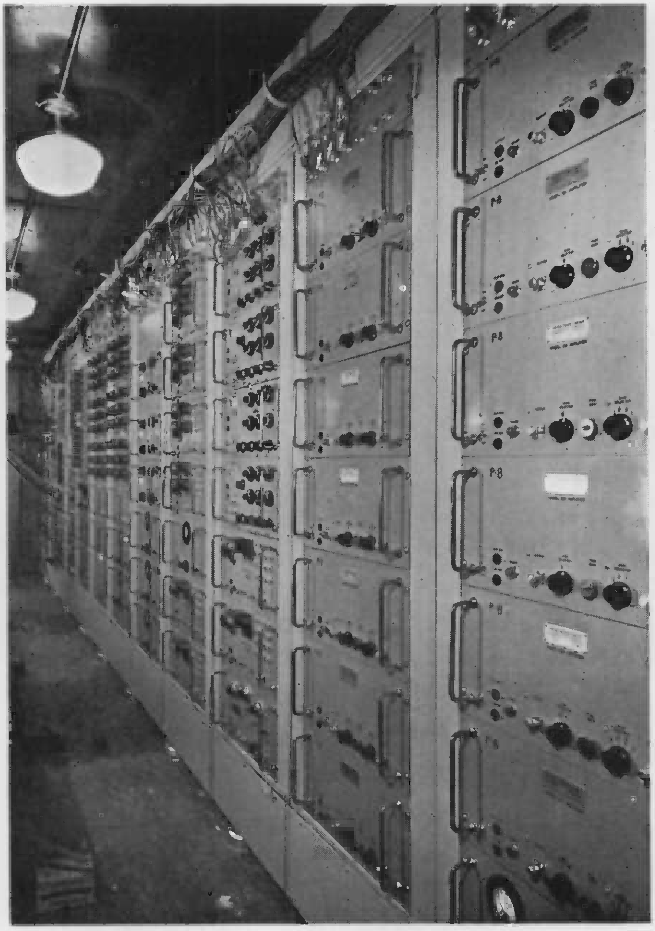

Fig. 7. Inside view of electronics van showing equipment required to select and record neutrino signals.

The liquid was viewed from the slab ends by 110, 5 in. photomultiplier tubes, 55 on each detector end. This large number was to obtain the required light collection and hence energy resolution. In order to make the detector response reasonably independent of the origin of the scintillation light, the photomultiplier tubes were separated from the scintillating liquid by means of a transparent window. A light match was provided by using non-scintillating TEB in the end sections. This arrangement made for a uniformity of light collection better than 10%. Figure 5 shows an inside view of one of the detector tanks. Figure 6 is an end view of the detector array inside a massive lead shield. Figure 7 shows the electronics van complete with the electronics required to sort, select, and record the various time and spatial coincidences of pulses resulting from reaction (1) and the above detector arrangement.

II. GAMMA-RAY DETECTION

In view of the extreme sensitivity of the detector and the relative infrequency of neutrino events expected, we became concerned about the backgrounds due to the detector itself. Since the detector was the most sensitive gamma counter available, it occurred to us to build a cylindrical insert about 23 cm in diameter, 75 cm high and check the gamma activity of construction materials. R. L. Schuch of Dr. Langham's group suggested making a larger insert—a 51 cm diameter seemed sufficient—and putting a person in it. That seemed like a very good idea, and we did,[5] as shown in Fig. 8. Appropriately enough, Dr. Langham was the first person to be counted in this fashion. We also persuaded him to carry with him into the counter a 0.1 μc radium source. We found that this amount of activity counted at the rate of ~1000 counts/sec and was some fifty times the readily detectable K^{40} content of the body. Since then (1953), as you have heard in detail from previous

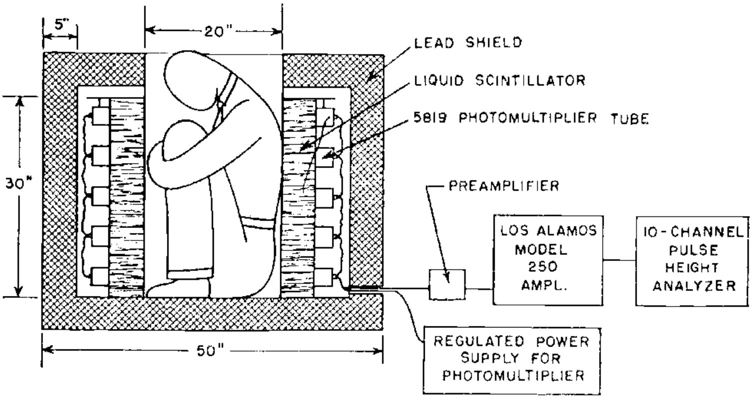

Fig. 8. Schematic view of first human counter (1953).

speakers in this conference, this total gamma absorption use of large volume liquid scintillation detectors has been very successfully exploited.

III. APPLICATIONS TO NEUTRON DETECTION

A system with high neutron efficiency can be used for neutron studies in many ways. Consider a hydrogenous medium which is essentially infinite from the point of view of a neutron and cause a fast neutron to be liberated within this medium. The neutron recoils and slows down in billiard ball type collisions pursuing a tortuous path, and eventually, if the medium contains only carbon and hydrogen is captured by a proton producing a 2.2 MeV capture gamma-ray. Our liquid scintillation detectors are indeed quite large relative to the mean free path for a neutron–proton collision ($\lambda = 20$ cm for a 10 MeV neutron in toluene) and the detection efficiency insofar as containing the neutron is concerned is consequently near 100%. On the other hand if the capture gamma-ray is to be absorbed in the scintillator the system has to be somewhat larger because of the greater absorption mean free path for gamma-rays. These considerations make for a system which for efficient detection of high energy neutrons measures ~ 1 m across. Now why does one care about this capture gamma in the first place when the recoil proton can be detected? The reason is that if the time delay between the initial slowing down pulse of the neutron and the neutron capture as signaled by the capture gamma can be used to obtain a delayed coincidence, the background will be greatly reduced. Furthermore, the distribution of time intervals can be very useful if you are interested, for example, in measuring the number of neutrons simultaneously released in a given event, and do not care particularly about their energy.

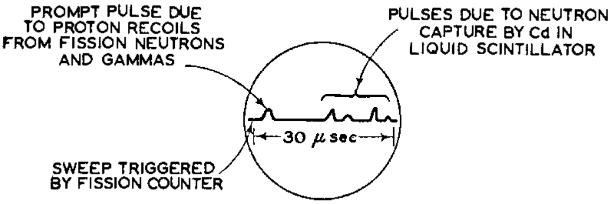

Fig. 9. Sketch of oscilloscope sweep showing multiple neutrons from fission. Sweep is triggered by a signal from a fission counter located inside the large volume scintillation detector.

III.1 *Fission neutrons*

Suppose we have an element such as Cf^{152} which undergoes spontaneous fission located in a fission counter which is placed in the center of our large scintillation detector. If we trigger an oscilloscope sweep when a fission occurs, we can then observe the number of fission neutrons associated with that particular fission as subsequent pulses on the ensuing trace as shown in Fig. 9.

The probability that more than one neutron would be captured within the resolving time of the system (0.2 μsec) and so result in a miscount is very small for a mean capture time fixed at $\sim$ 10 μsec. The mean capture time is determined by the amount of cadmium compound dissolved in the scintillator.*† Fission experiments of this type have been done at Los Alamos and Berkeley. Thc motivation for such an experiment goes something as follows. Quite apart from the interest in the number of neutrons which come from the fission process because of its economic importance, there is interest on the part of physicists in the details of the fission process. It is known that there are many modes of fission and that the neutron yield in a given fission is related to the ratio of the fission fragment masses, a ratio which determines for a given initial nucleus the excitation of the fragments. With these large scintillation detectors it has become possible as Whetstone *et al.* and others at Los Alamos have demonstrated[6] to study this relationship directly. DIVEN *et al.*, at Los Alamos,[7] and HICKS, *et al.* at Berkeley[8] studied earlier the variation in neutron numbers from fission without, however, relating this information to the fission mode.

III.2 *Capture neutrons*

Another yet only partially exploited use of these large detectors is in the measurement of neutrons emitted by nuclei which have captured negative mu mesons or muons (μ^-). You will recall that although muons interact very weakly with nuclei, negative muons have been observed to produce neutrons and nuclear disruptions via such interactions. The reason a μ^- can interact despite this weak coupling to the nucleus is that it forms an atom much as a negative electron does but is more tightly bound, i.e. is closer to the nucleus. This proximity to the nucleus greatly enhances the probability of absorption by the nucleus. When a negative muon undergoes nuclear absorption a neutrino is emitted taking off most of the energy but enough excitation remains with the nucleus to 'boil off' a few neutrons. Until recently, rather cumbersome techniques using relatively inefficient systems were employed to study this neutron boil off. Several months ago the Berkeley group reported the use of these large liquid scintillation detectors in conjunction with cosmic ray μ^- to measure neutron multiplicities from mu meson capture.[9] The elementary reaction is

$$\mu^- + \mathrm{p} \rightarrow \mathrm{n} + \nu_+ \qquad (2)$$

where p is to be understood as a proton in a nucleus.

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* This application was first suggested by Dr. H. Karr of Los Alamos.

† A paper by Ronzio, Cowan and Reines, *Rev, Sci. Instrun.* **29**, 146 (1958), describes cadmium octoate which we have found to be superior to the cadmium propionate first employed because of its chemical stability, less corrosive character and greater solubility.

III.3 μ^- *Fission neutrons**

Another application of these techniques that I would like to propose is to study fission induced by negative muons. Electronuclear machines could provide a beam of muons although given a large enough lump of target material, cosmic rays might suffice as a meson source.

The fact that one can, in some cases, use as sources, not machines but the cosmic rays, makes possible some meson research for people who do not have machines available to them.

III.4 μ^- *Catalysis*

Another example of a possible application of these counters to neutron detection is in the study of mu meson catalysis of light particle nuclear reactions. You may have heard how Canadian uranium stocks fell several months ago when a newspaper carried the announcement that uranium was now unnecessary because of the discovery of 'cold fusion'. The physics behind this uncalled for optimism was the discovery by the Berkeley group of Alvarez[10, 11] that the mu meson could bind together as a molecule two hydrogen nuclei and hold them together closely enough for them to penetrate the coulomb barrier, reacting nuclearly. It then was liberated making possible the catalysis of a second reaction. This catalytic action of the μ^- meson was discovered by an ingenious analysis of tracks seen in a liquid hydrogen bubble chamber. It turns out that because of the high neutron sensitivity of large scintillation detectors they can be designed to study this phenomenon. The experiment has not been done as yet, but a certain amount of preliminary planning at Los Alamos indicates it to be a promising approach to the problem of mu catalysis studies. Consider, for example the reaction catalyzed by μ^-

$$\mu^- + \mathrm{d} + \mathrm{t} \rightarrow \mathrm{He}^4 + \mathrm{n} + \mu^-. \tag{3}$$

In this case a neutron is produced and the μ^- is left free to catalyze another reaction. This process repeats until the μ^- is captured by a He^4 nucleus or decays with the emission of an electron and two neutrinos according to the reaction

$$\mu^- \rightarrow \beta^- + v_+ + v_- \tag{4}$$

$$\text{half life } \tau = 2 \times 10^{-6} \text{ sec}$$

By observing the entry of a μ^- into the system and then detecting the multiplicity of resultant neutrons, the length of the catalysis chain can be determined. Because of the moderation time of the neutrons prior to capture

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* The possibility that μ^- might induce fission was proposed by J. A. WHEELER as a result of the energy released in the orbital capture of μ^- by a heavy nucleus such as uranium or in the nuclear capture of the μ^-: *Rev. Mod. Phys.* **21**, 133 (1949). Experiments by Galbraith and Whitehouse (1953) reviewed by R. SARD in *Progress in Cosmic Ray Physics* Vol. II, Interscience Publishers (1954) have as yet been unsuccessful in observing μ^- induced fission. These workers concluded that $<1/4$ of the μ^- stopped in uranium, induced fission.

(~10 μsec) one would see the μ^- decay electron pulse before the neutron capture pulses. An oscilloscope record of the event might be as shown in Fig. 10. A sketch of a possible experimental arrangement is shown in Fig. 11.

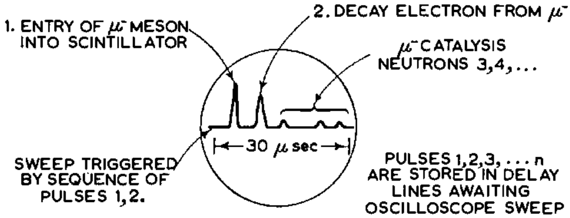

Fig. 10. Possible oscilloscope record in a μ^- catalysis experiment. The first pulse is due to the entry of a μ^- meson, the second to μ^- decay, and the subsequent pulses to capture of the neutrons produced in the catalysis chain.

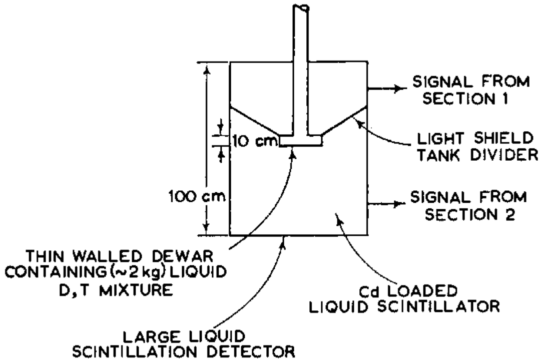

Fig. 11 (a).

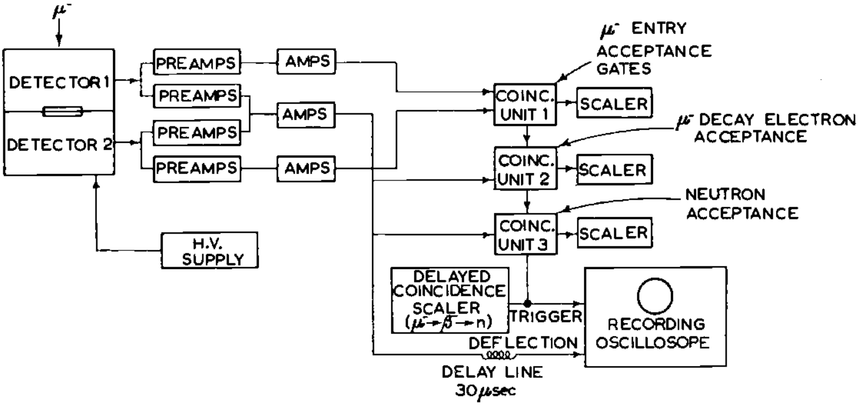

Fig. 11 (b).

Fig. 11. a, b. Schematic of an experimental arrangement proposed for the study of μ^- catalysis.

It should be mentioned that it is not yet theoretically clear how many reactions a given μ^- will catalyze and indeed that would be the main point of such an experiment as described here.

III.5 *Measurement of neutron energy*

The statement is often made that neutron energies cannot be measured using liquid scintillators because of the nonlinear response of the scintillator to recoil protons, and I should also add, because of the collisions with carbon which occur, effectively adding to the nonlinearity.[12] This statement is true except in the case of monoenergetic neutrons such as one could obtain at a fixed angle from protons incident on a lithium target. In this instance let us assume that n, ($\sim$ 10) neutrons entered the detector in one pulse. Then the number of neutrons, n, associated with the recoil pulses would be given by the sequence of pulses indicating neutron capture. The height of the recoil pulse, E_n, divided by the number of neutrons, n, would for large n be a unique function of the neutron energy, E, and

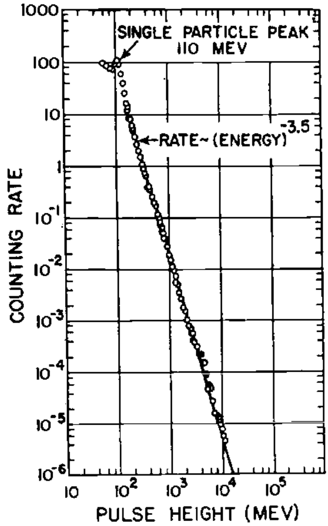

Fig. 12. Cosmic ray energy spectrum at Los Alamos (7200 ft) as seen by the 300 liter scintillation detector of Fig. 2. Pulses of energy $> >100$ MeV, the mean energy deposition of a minimum ionizing particle, are due to multiple particles in the detector either from externally produced showers or perhaps, although less likely, from showers occurring in the detector itself. A fraction of these very high energy pulses is due to photomultiplier tube and socket noise which was not eliminated by requiring coincidences in this experiment.

$$E = f\left(\frac{E_n}{n}\right) \tag{5}$$

IV. STABILITY OF THE NUCLEON

An unusual application of large scintillation detectors has been made in connection with a determination of the stability of a nucleon (proton or neutron in a nucleus) against decay into lighter particles. It is assumed in nuclear physics, that a nucleon is stable against such decay and indeed the principle of nucleon conservation is one of the cornerstones of the subject. One can make arguments about the stability based on the level of ambient radioactivity leading to such lifetimes as $> 10^{17}$ years and such a limit is consistent with the behavior of nuclei. There is, however, no law which prohibits a nucleon from decaying with a lifetime greater than this. It turns out that the large liquid scintillation detector is made to order for such a determination. The idea of the experiment was to locate the detector underground so as to shield it from cosmic rays. We then observed the counting rate in the energy range to which products of nucleon decay might be expected to contribute. This rate was then related to the number of nucleons in the detector and vicinity. In this manner we were able to set a lower limit on the nucleon lifetime of 10^{22} years.[13]*

V. THE LARGEST DETECTOR

It is interesting to inquire about the limits in size on a liquid scintillation detector. Without at the moment asking why one would want the biggest one possible, how large could one be made? A primary limitation is imposed by the absorption mean free path for the scintillation light. It seems technically feasible to make liquid with an absorption path in excess of 10 m. Therefore we take as an approachable limit a giant detector measuring 10 m across. A cylinder 10 m across and 10 m high would have a capacity of 600 tons of scintillator! For a light collection efficiency of 1 % of the photons produced in the liquid 1000, 5 in. photomultiplier tubes would be required, assuming a non-reflecting tank wall. Use of a white wall should reduce this number to a few hundred in which case 1 MeV should be detectable with an energy resolution of something like ± 50%.† The time response of such a detector would be limited by the transit time of light across it and so, allowing for two transits, would be about 0.1 μsec.

It would be nice to have compelling reasons to build such a colossal

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* Recent work by Reines, Cowan and Kruse, *Phys. Rev.* **109**, 609 (1958), raises this limit considerably.

† The energy resolution is here due primarily to statistical fluctuations in the number of photoelectrons produced in the photomultiplier tubes. If we take 150 eV energy loss in the scintillator to be required per photon emitted and 10% to be the photoelectric efficiency of the photocathode then 1000 phototubes would have liberated in them 10 photoelectrons. This number is raised by the wall reflectivity. The statistical fluctuation is then $\pm 100/\sqrt{10} = \pm 32\%$.

detector, but none have yet been advanced which seem to justify the effort and expense. However, let us think about it a little. A detector this size has as much mass per unit area as the entire atmosphere: it represents a total absorber for charged particles of energies up to 1.5 BeV.* In it one could observe the process of shower production due to high energy particles much like those produced by high energy cosmic rays—but with one essential difference. The time of traversal of the various particles through the detector is so small due to its size that the unstability of some of the particles involved, i.e. π and μ mesons plays no essential role in the showers developing in the giant counter as they do in showers produced in the extended earth's atmosphere. The selection of the primary particle could be effected by means of a Čerenkov counter and insurance against the simultaneous entrance of more than one energetic particle could be gained by means of large anti-coincidence scintillation detectors. The shower production could be enhanced by means of plates made of lead or other materials placed in the detector. It would even be possible to study associated neutrons by means of delayed coincidences.

There is another side to this ultra-colossal business and that is the emergence of very high energy machines. Particles made in such multibillion electron-volt machines have many meters of range and one could for example study various shower phenomena induced by these energetic particles using such large detectors. It is not possible to study these complicated many-particle processes over a large number of generations by other techniques although very large liquid bubble chambers such as the one currently under construction at Berkeley by the Alvarez group may approach the task† if lead plates are incorporated. Some hint of this application is given by Fig. 10 which shows the energy spectrum seen by a 300l. scintillation detector exposed to cosmic rays. Since the maximum energy which can be deposited by a minimum ionizing ray passing through the detector is $\sim$ 100 MeV, higher energy pulses are presumably due to multiple particles in the detector either from an externally produced shower or perhaps less likely, from a shower which occurs in the detector itself.

VI. CONCLUSION

We have seen several examples which indicate that the very large volume liquid scintillation detector has already found important uses in neutrino research, charged particle detection, gamma-ray detection, and neutron counting. It seems reasonable to expect that many more applications of these

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* We are here considering only ionization loss. In fact the bremsstrahlung losses for electrons and μ-mesons and nuclear interactions for nucleons increases the number manyfold.

† The fundamental processes are of course much better studied with photographic emulsions and bubble chambers. Counting experiments are most useful when one knows what one is looking for.

techniques will be found and that the field of large volume liquid scintillation counting will continue to merit investigation for a long time to come.

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PART VI

DEVELOPMENTS IN FOREIGN LABORATORIES