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A PROPOSED EXPERIMENTAL TEST OF THE NEUTRINO THEORY

Luis W. Alvarez

April 18, 1949

Berkeley, California

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A PROPOSED EXPERIMENTAL TEST OF THE NEUTRINO THEORY

I. Introduction

The experiment outlined in this proposal has the possibility of giving an answer to the important question, "Does the neutrino exist?" It is unfortunate that at the present time, there is no convincing experimental proof that neutrinos exist. Two recent articles[x] review the status of various experiments which could give information about neutrinos. In general, these experiments give results in agreement with the predictions of beta decay theory. But actually, if even the most complete of the "recoil type" experiments could be performed satisfactorily, all that could be concluded would be the following: The energy and momentum relationships in beta decay are consistent with the theory that the known energy deficit is carried away by a single particle. But to emphasize the fact that this would not constitute a proof of the real existence of that particle, the following quotations from the review articles should be noted. Crane says, "All of the evidence about the neutrino is, as already pointed out, indirect in character, since neutrinos have not yet been caught after leaving the nucleus.....It can, of course, be argued on very general grounds that, if energy is not conserved between nucleus and electron, momentum should not be expected to be conserved either; and in consequence of this, it has often been remarked that the recoil experiments add nothing that is really new to our knowledge....." Allen concludes his article by saying, "Practically all the experimental evidence indicates that there is an apparent non-conservation of momentum in the beta decay process, and that the neutrino hypothesis is at least one explanation of the missing momentum." (Underlining added.)

[x] J. S. Allen, Am. Jour. Physics, 16, 451, (1948)
H. R. Crane, Rev. Mod. Phys. 20, 278, (1948)

It is instructive to compare our present thoughts about the neutrino with those of an earlier generation of physicists concerning the ether. There were probably

no competent physicists in 1890 who doubted the existence of the ether; physicists today write and speak about the neutrino as though it has a real existence, but they have an intellectual reservation about the validity of the neutrino hypothesis, which their predecessors apparently did not have about the ether. (It is interesting to read the literature of 1900 and see that all of the explanations of the Michelson-Morley experiment were in terms of an ether theory.)

It is therefore important that at least one experiment be performed in which neutrinos are made to do something after they have left the nucleus.

It was pointed out by Bethe in 1936, that there is one type of nuclear process which a neutrino will certainly excite. (In the rest of this paper, the existence of the neutrino will be assumed, for purposes of discussing and calculation, and the usual reservations will not be made explicitly.) Since the neutrino is emitted during beta decay, it must be able to reverse the process, and the cross section for the inverse reaction may be calculated from the principle of detailed balancing. The fact that the cross section may be calculated from statistical mechanical considerations of the most general character, is what makes the proposed experiment a crucial one for the neutrino theory. The process of interest in the proposed experiment is the "inverse electron capture." According to the theory of electron capture, a neutrino is emitted when an electron is captured by a radioactive nucleus. The inverse process involves the capture of a neutrino by a stable nucleus, and the emission of an electron. (According to one theory of β-decay, the distinction between neutrinos and anti-neutrinos could be of importance, since anti-neutrinos would be required to reverse an electron capture process, while a pile emits ordinary neutrinos. Reasons for believing that pile neutrinos will be capable of reversing an electron capture reaction are given in Appendix I.)

The experiment involves the exposure of a stable substance, Cl^{37}, to the neutrinos from a pile. After the irradiation, the radioactive isotope, A^{37}, would be

separated from the original material, and its activity measured. A^{37} is a known electron-capturing isotope, whose half life is 34.1 days. The neutrinos would excite the nuclear reaction:

$$Cl^{37} + \nu \rightarrow A^{37} + e^- \qquad (1)$$

The observed activity would correspond to the reaction:

$$A^{37} + e^- \rightarrow Cl^{37} + \nu \qquad (2)$$

The reason for choosing this reaction will become evident when the order of magnitude of the expected activity is discussed.

In theory, the experiment is as simple as any involving the production of a radioactive isotope by irradiation of a stable material with a flux of particles. It is straightforward to calculate the activity produced, if the incident flux, the cross section for the reaction, and the mass of the bombarded material are known. The difficulty of the proposed experiment comes entirely from the smallness of the cross section, which is of the order of 10^{-45} cm^2. (The average cross section for pile neutrinos is shown in Appendix II to be 2×10^{-45} cm^2.) An idea of the magnitude of this cross section can be had from the fact that the probability of a neutrino being captured in passing straight through the earth is only about one in 10^{12}. It is not surprising that it has generally been felt that this effect was too small to be observed.

Since the construction of atomic piles has provided neutrino sources of very great intensity, a number of persons have independently looked at the problem in recent years. Pontecorvo[x], at Chalk River, has published a lecture he gave on the

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[x] B. Pontecorvo, declassified Canadian report issued by AEC, Oct. 8, 1949. (Lecture given Nov. 20, 1946.) PD-205

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subject of inverse β-decay, including some data on the same reaction considered in this proposal. He made an estimate of the $Cl^{37} + \nu \rightarrow A^{37} + e^-$ cross section, by

an order of magnitude type calculation. (Bethe had made an exact calculation on the basis of the now-discarded K.U. theory, in 1936.) Unfortunately the numerical factors, which cannot be evaluated in a derivation of the type employed by Pontecorvo, are quite unfavorable, and his cross section was overestimated by a large factor.

It is worth noting in this connection that there are two different ways of looking at this experiment. Pontecorvo, who is working in a laboratory equipped with a pile, was originally going ahead with plans to look for the inverse electron capture effect, and if it were unobservable, to set a new upper limit to the cross section for a neutrino effect. (Recent reports from Chalk River indicate that these plans have been abandoned.) Determinations of upper limits have been made in the past, and have yielded the following values: Three quite different experiments by Nahmias, Wollan and Crane have shown that the interaction cross section of neutrinos with atoms is less than 10^{-30} cm^2. Crane uses geophysical evidence concerning the rate of production of heat in the earth, by neutrinos from the sun, to show that cross sections in the range 10^{-32} to 10^{-36} or 10^{-37} are also excluded. He points out that the possible range between 10^{-30} and 10^{-32} may easily be excluded by experiments with chain reacting piles, and probably will be in the near future. Certainly, Pontecorvo's experiment would have done that, as well as push the limit to perhaps 10^{-41}, if it really is of the expected magnitude of 10^{-45}. (This is on the assumption that his experiment was to be done on the scale outlined in his report: one cubic meter of CCl_4 and a counting rate of 1 per minute.)

Although it would be important to know that the cross section is less than 10^{-41} cm^2, nothing could be concluded about the existence of the neutrino from such information. However, if it could be shown that the cross section were less than 10^{-45} cm^2, the whole neutrino theory would have to be re-examined critically, and it is quite possible that the theory would have to be discarded. If, on the

other hand, a cross section of this magnitude were observed, it would prove conclusively that neutrinos had a real existence. The philosophy behind the proposed experiment is that every effort should be made to increase the sensitivity to the point where the theoretical cross section would yield an effect many times the expected background. One could, of course, increase the effect arbitrarily by irradiating larger masses of material, but the really important consideration is the ratio of effect to background. In his preliminary report, Pontecorvo merely states that the background due to cosmic rays would be "very small;" this statement would be correct if the cross section were 10^{-42}, as he estimates, and if the neutrino flux of 10^{14}, which he looked forward to having available in the future, were used.

It will be shown later in this proposal, that if the experiment is to be done with presently available neutrino sources, the most important experimental problems lie in the elimination of the various types of background. The counting rates due to the neutrino induced activities are quite adequate to give a high statistical accuracy in a measurement of the cross section, assuming an absence of background effects. But if no serious efforts were made to eliminate the background, the expected activity would be "lost in the background."

II. Experimental Procedure

The saturation counting rate, A, of a sample prepared by the bombardment of N atoms, each with cross section σ, in a flux of nv particles per cm^2 second, is

$$A = nv \times N\sigma \qquad (3)$$

According to Appendix III, the neutrino flux at a distance D feet from the center of a pile operating at $P \times 10^8$ watts, is

$$nv = \frac{1.70 \times 10^{15} P}{D^2} \qquad (4)$$

If one takes "reasonable values" of P, D, and N, and uses them to evaluate A, he finds that A is vanishingly small. D must be greater than the distance from the

outside of the shield to the center of the pile, plus half the thickness of the irradiated material, plus several extra feet of shielding needed to reduce the flux of fast neutrons. (The background traceable to fast neutron effects is discussed in Appendix VI.) D is not really an adjustable constant, since it must be in the neighborhood of 40 feet. The power P is of course determined by the pile available. Sample activities will be tabulated for powers of 0.3, 1, and 3 x 10^8 watts. One therefore has only N as an adjustable constant, and this must be made as large as practicable. Since CCl_4 is the most attractive chlorine compound, and since it is available in tank car lots, N is chosen as the number of Cl^{37} nuclei in one tank car of CCl_4. Even with such large quantities of material to be irradiated, the activity is exceedingly small by ordinary standards. However, it will be shown that these activities are quite adequate to make a precise measurement of the neutrino capture cross section.

A standard tank car holds about 40 metric tons of CCl_4 with molecular weight = 153.8. Cl^{37} is 24.6°/o abundant, so

$$N = \frac{4 \times 10^7}{153.8} \times 6.03 \times 10^{23} \times 4 \times 0.246$$

$$N = 1.57 \times 10^{29} \text{ atoms of } Cl^{37}$$

If we take D = 35 feet, the saturation counting rate of A^{37} is

$$A = \frac{1.70 \times 10^{15}\ P \times 1.57 \times 10^{29}}{(35)^2} \times 20.4 \times 10^{-46}$$

$$A = 4.35 \times 10^{-4}\ P \text{ counts per second}$$

$$A_\infty = 37.5\ P \text{ counts per day}$$

For an irradiation of two half lives (68 days) the activity would be

$$A_2 = 0.75 \times 37.5P$$

$$\boxed{A_2 = 28\ P \text{ counts per day}} \qquad (5)$$

Before discussing the experimental methods which make possible the determination of such small activities, it should be instructive to plot the decay curves of an activity of this magnitude, together with the probable errors, to show that the numbers are large enough to give reliable information about the capture process. Figure 1 shows theoretical decay curves of the A^{37} activity, for three values of P, with points every ten days. (These values are chosen to show that for all powers much less than 30 megawatts, the background effect is too large, while for all powers greater than 300 megawatts, the background is quite negligible.) The background due to the counter is assumed to be one per day. (Methods of achieving such a small background are described in Appendix II.) It is evident from an inspection of these curves that reliable measurements of the A^{37} activity could be made with piles of the three powers assumed.

But it must be noted that there are three other processes which lead to the production of A^{37} in a tank of CCl_4. Protons will give the reaction

$$Cl^{37} + p \rightarrow A^{37} + n \qquad (6)$$

The three sources of protons are: (1) cosmic rays, (2) n,p reactions in the CCl_4, from fast neutrons which leak through the pile shielding, and (3) α,p reactions on chlorine, from α-particle emitting impurities in the CCl_4. These three sources of background A^{37} are discussed in Appendices IV, V and VI. The cosmic ray effect is the most difficult to eliminate, and in his earliest evaluations of this effect (January 1948), the author could find no method of eliminating it. The background due to such a process is approximately 10^4 counts per day at sea level, and experiments of Perkins showed that the mass absorption coefficient of the proton-producing cosmic radiation was very small in lead, but much larger in air. This indicated that the radiation was unstable, and that the apparent absorption coefficient in air was due to the decay process. On this basis,

shielding would be ineffective, and for that reason, the author concluded that the experiment was not feasible. In March 1949, Perkins published new data on the absorption of the star-producing radiation in ice, which showed it to be close to that of air on a mass basis. These new data show that the star-producing radiation is actually absorbed primarily by nuclear encounters, and make it possible to consider the process of shielding.

In order to reduce the cosmic ray induced A^{37} activity to about one per day, it is necessary to have a thickness of shielding equal to 9 mean free paths. This amounts to 60 feet of water, or about 40 feet of concrete. Two separate methods of shielding suggest themselves: (1) the tank of CCl_4 could be placed in a tunnel under the pile, or (2) a shield of water, dirt, or concrete could be built over the tank, which would be set on the ground level, close to the pile. In view of the great expense involved in the second method of shielding, only the first will be considered.

The problems involved in the extraction of minute amounts of A^{37} from many tons of CCl_4 are discussed in Appendix VIII. Similar separations of noble gasses have been done on the laboratory scale for years, and are done in a routine fashion daily in hospitals all over the world. Radium is often kept in solution, and the radon gas is extracted and introduced into small glass or gold "needles," for therapeutic purposes. Radioactive Krypton and Xenon can be extracted from neutron irradiated uranium solutions, with a high degree of efficiency. The main problems connected with the extraction of A^{37} in tracer amounts from tons of CCl_4 are those of a chemical engineering nature, and can no doubt be solved without great difficulty. Helium can be used as the "carrying gas," to sweep the A^{37} from the CCl_4. The separation of A^{37} from He^4 is easily accomplished by passing the gas through active charcoal cooled to liquid air temperature. Finally, the charcoal would be warmed and enough ordinary argon, neon or helium would be introduced into the system

to sweep out the A^{37}, and act as the counting gas of a small proportional counter.

The A^{37} counts are due to Auger electrons, which follow the emptying of the K shell in K-electron capture. In heavy elements, a quantum of K-X-rays is usually emitted when an empty K-shell is refilled. But this happens in only 7°/o of the A^{37} cases; in the remaining 93°/o, the K-excitation energy is given to an outer electron, in a sort of internal conversion, or Auger, process. The Auger electrons have an energy of 2.8 Kev, and a range of approximately 0.15 mm in He at atmospheric pressure. This very small value of the range is what makes possible the attainment of backgrounds of the order of one per day in a proportional counter. Spurious counts from α-particle impurities in the walls will be approximately 1 per day, and since each α-particle makes hundreds of times as many ions in the counter, such counts may easily be eliminated by a discrimination circuit. The net α-particle background should be of the order of 0.02 count per day. β- and γ-ray counts, and cosmic ray meson counts are eliminated by an anti-coincidence shield counter. Since the A^{37} counter will be 3 mm in diameter, and 1 cm long, the shield counter can easily surround it completely. Although other experimenters have used shield counters to reduce background effects, their published backgrounds are very much greater than the assumed value of one per day. The reason that such large factors of improvement can be made when using A^{37} comes entirely from the exceedingly short range of the Auger electrons. If one is trying to count C^{14} electrons with an anti-coincidence shield counter, as Libby has recently done, he must make his counter walls thick enough to keep the desired electrons from reaching the shield counter. But in so doing, he increases the probability that β-rays from radioactive impurities in the counter walls will give a count in the inner counter, from a β-ray which cannot penetrate into the shield counter and so be eliminated. When counting A^{37} electrons, one can make the walls as thin as technically feasible, which is of the order of a few milligrams per cm^2. This gives two benefits; the

amount of radioactive contamination in the walls is cut down, and the probability that any contamination β-ray reaches the shield counter is increased. Numerical examples are given in Appendix VII, to show that it should not be difficult to achieve a background of 1 count per day.

Now that the general outlines of the proposed experiment have been set down, it is possible to justify the choice of A^{37} as the radioactive substance. An examination of the isotope table shows that no other substance combines all the highly desirable features of A^{37}, and in fact, no other isotope even comes close to being a worthwhile candidate for the experiment. The desirable properties are:

1. The radioactive isotope should decay primarily by emitting very short-range electrons.

2. It should exist in a gaseous molecule, and preferably be a noble gas.

3. The substance from which it is produced by neutrino capture should be available in liquid form, in large quantities.

4. The mass difference between the initial and final nucleus should be small, and known. (As will be shown in Appendix II, the cross section falls rapidly as this mass difference is increased.) Points 4 and 1 require that the active substance decay by electron capture.

5. The half life should be long, so that the probable errors of the counting rate may be kept low. This point is fortunately consistent with Point 4.

6. The decay should be allowed, according to the terminology of β-theory. Although A^{37} is probably an allowed transition, it is not possible to say so with certainty. The only effect this could have on the experiment is that the theoretical cross section could be somewhat larger than the quoted value of 2.0×10^{-45}. This point is discussed in Appendix II.

APPENDIX I

Equivalence of Neutrinos and Anti-Neutrinos

According to the usual formulation of the Fermi theory of beta decay, a nucleus undergoing negative beta decay emits an electron and a neutrino, and at the same time, one of its neutrons is changed into a proton. In positron decay, an anti-neutrino accompanies the positive electron, and in electron capture, an anti-neutrino of definite energy is given off after the electron is captured. Neutrinos and anti-neutrinos are both considered to have the same small mass (probably zero), spin 1/2, and zero charge. Since they probably have no magnetic moment, it is hard to see in what physical way they might differ. But from a purely formal viewpoint, they would be expected to annihilate each other under the proper conditions, giving rise to two gamma rays. For most purposes, the formal distinction is ignored, and one seldom sees references to anti-neutrinos in the literature of beta theory.

It is important to know if there is any experimental reason to believe that neutrinos are really equivalent to anti-neutrinos. If the proposed experiment gave a negative result, i.e. did not show the expected A^{37} radioactivity, it could presumably be concluded that: (1) neutrinos did not exist, or (2) neutrinos and anti-neutrinos may exist, but if so, they differ in their ability to reverse positive and negative beta processes. If possibility (2) could be eliminated, then the experiment would have the crucial nature that would make it of greater interest.

Majorana has proposed a modified Fermi theory which involves the concept of the equivalence of neutrino and anti-neutrino. Until recently, there was no experimental way to distinguish between the original Fermi theory and that of Majorana. Within the past few months, however, an experiment performed by Fireman[x]

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[x] E. L. Fireman, Phys. Rev. 75, 323 (1949)

has given very strong evidence that the Majorana modification is correct. Fireman has made a search for the so-called "double beta decay," and has found that Sn^{124} does transform into Te^{124} by the simultaneous emission of two beta particles. The intermediate isobar, Sb^{124} is heavier than either of its neighbors, so the decay could not occur in two stages.

The importance of the discovery of double beta decay to the neutrino - anti-neutrino question comes from a measurement of the half life of the process. Fireman quotes this as being in the range $4 - 9 \times 10^{15}$ years. The Majorana theory predicts lifetimes in the range 10^{14} to 10^{16} years, depending on the mass difference between Sn^{124} and Te^{124}. However, the original Fermi theory predicts lifetimes approximately 10^{10} times longer. The reason for the difference in the prediction of the two theories is easy to understand in a qualitative manner. According to the Fermi theory, a neutrino must be emitted whenever a beta ray is given off by a nucleus. The emission of a neutrino is of course equivalent to the absorption of an anti-neutrino. If one keeps the distinction between the two types of neutrinos, two neutrinos must be emitted in the Fireman process. But if the two types of particles are equivalent, a double beta decay can be accompanied by the virtual emission of a neutrino, and its subsequent reabsorption. The difference in the two calculated half lives comes directly from the volumes in phase space available in the two cases.

On the basis of Fireman's work, one may conclude that there is no real difference between the two types of neutrinos. Specifically, one may interpret his experiment as showing that the reabsorption of the virtually emitted neutrino (which accompanied the emission of a negative beta particle), was responsible for reversing an electron capture process, i.e. giving rise to the emission of a negative electron. Since this is just the sequence of events which is envisaged as occurring in the proposed experiment, there can be no doubt (assuming the correctness of Fireman's difficult experiment), that pile neutrinos are capable of reversing an electron capture process.

APPENDIX II

Neutrino Cross Sections

There are several steps involved in the calculation of the average pile neutrino cross section for the $Cl^{37} \longrightarrow A^{37}$ reaction. From arguments similar to the principle of detailed balancing, one may evaluate the cross section as a function of individual neutrino energy. Since the neutrino energies considered in the capture process are not identical to those of the neutrinos emitted in the decay, the detailed balancing argument cannot be used in a precise sense, but an extension of it, which involves an assumption as to the energy variation of the matrix element, is involved. For allowed transitions, it is supposed that the matrix element is independent of energy.

The primary cross section function is then averaged over the neutrino distribution for arbitrary values of the "upper limit." This operation gives the average neutrino cross section as a function of the upper limit of the neutrinos (or β-rays) emitted by a particular radioactive substance. Finally this new cross section function must be averaged over the distribution of upper limits among the fission products. This operation gives the average neutrino cross section for all pile neutrinos.

The first of these three steps has been done independently by three of the author's colleagues, to whom he is greatly indebted. The results of their work are identical. The formula was derived by B. A. Jacobsohn, L. I. Schiff, and M. Lampert. The derivation given below was supplied by Dr. Schiff.

The "forward reaction" is

$$A^{37} + e^{-} \longrightarrow Cl^{37} + \nu$$

The rate of K-capture is

$$\frac{1}{\tau} = 2 \times \frac{2\pi}{\hbar} \left| H_F' \right|^2 \rho$$

where the factor 2 accounts for the 2 K electrons.

$$\rho = \frac{\text{number of neutrino states}}{\text{energy range}} = \frac{4\pi p^2 \left(\frac{dp}{dE}\right)\Omega}{8\pi^3 \hbar^3}$$

where Ω is the volume of the "box" in which the system is quantized. $p = E/c$, and the neutrino energy is $E = 1 + \Delta$, in units of mc^2. Δ is the nuclear mass difference between A^{37} and Cl^{37}, and $1 + \Delta$ is the atomic mass difference, in units of mc^2.

The matrix element is some constant times the product of the amplitudes of the normalized K electron and the neutrino wavefunctions.

$$H_F' = g\sqrt{\frac{Z^3}{\pi a_o^3}}\,\frac{1}{\sqrt{\Omega}}$$

Therefore

$$\frac{1}{\tau} = 2 \times \frac{2\pi}{\hbar} \times \frac{g^2 Z^3}{\pi \Omega a_o^3} \times \frac{(1+\Delta)^2 \Omega}{2\pi^2 \hbar^3 c^3}$$

$$\frac{1}{\tau} = \frac{2g^2}{\pi^2}\left(\frac{Z}{a_o}\right)^3 \times \frac{(1+\Delta)^2}{\hbar^4 c^3}$$

a_o is the "Bohr radius."

For the "reverse reaction,"

$$Cl^{37} + \nu \rightarrow A^{37} + e^-,$$

the transition rate is

$$\frac{\sigma_c}{\Omega} = \frac{2\pi}{\hbar}\left|H_R'\right|^2 \rho$$

where $\rho = \dfrac{\text{number of electron states}}{\text{energy range}} = \dfrac{4\pi p^2 \left(\frac{dp}{dE}\right)\Omega}{8\pi^3 \hbar^3}$

$$p = \frac{1}{c}\sqrt{(E-\Delta)^2 - 1}$$

$$\frac{dp}{dE} = \frac{E - \Delta}{c^2 p}$$

In this case,

$$H_R' = g \times \frac{1}{\sqrt{\Omega}} \times \frac{1}{\sqrt{\Omega}}$$

Therefore

$$\sigma = \frac{\Omega}{c} \times \frac{2\pi}{\hbar} \times \frac{g^2}{\Omega^2} \times \frac{\sqrt{(E - \Delta)^2 - 1}\,(E - \Delta)\Omega}{2\pi^2\hbar^3c^3}$$

$$\sigma = \frac{g^2(E - \Delta)\sqrt{(E - \Delta)^2 - 1}}{\pi\hbar^4c^4}$$

Eliminating the unknown constant g^2, we have,

$$\sigma^o = \frac{\pi}{2c}\left(\frac{a_o}{Z}\right)^3 \frac{1}{\tau} \frac{(E - \Delta)\sqrt{(E - \Delta)^2 - 1}}{(1 + \Delta)^2} \qquad \text{II-1}$$

The factors in the denominator evaluate the matrix element, in terms of experimentally measured quantities, and the energy factors in the numerator are the usual ones involving the phase space available for the ejected electrons. As was mentioned earlier, the assumption is made that the matrix elements for the two reactions are equal. If the transition $A^{37} \rightarrow Cl^{37}$ were allowed, this assumption should certainly be justified, since the matrix elements would then be approximately unity. If the transition were forbidden, the value of σ would be a lower limit. The calculations will all be done on the assumption that the transition is allowed, since it probably is, but the "bonus" to be had in the event that the transition is forbidden will be discussed later.

There is another possibility that the simple formula for the inverse cross section might be too low. (The fact that is really a lower limit would be of the greatest importance in the interpretation of the experiment, if no A^{37} were to be observed.) It is possible that neutrinos could excite transitions to be excited states of A^{37}, which would then decay by γ emission to the ground state. Since the final observation is of the total A^{37} activity, one must make an estimate of this

contribution to the cross section. This is done in this appendix, and it is very doubtful that a significant increase in the total neutrino cross section will result from the existence of these higher states in A^{37}. If a higher state is to contribute to the cross section, it must have approximately the same matrix element as the (allowed) ground to ground transition. In other words, the product of the factors in the denominator of Schiff's formula will have the same value for any contributing state. The cross sections for the ground state and the excited states will differ only in the energy factors in the numerator, through the energy Δ.

From the formula for $\sigma^{o}(E,\Delta)$, we may calculate $\sigma(E_M, \Delta)$, which is the average cross section for all the neutrinos from a β-emitter of upper limit E_M.

$$\sigma(E_M, \Delta) = \int_{1+\Delta}^{E_M} \sigma^{o}(E, \Delta) f(E) dE \qquad \text{II-2}$$

where f(E) is the distribution in energy of the neutrinos from a β-emitter of upper limit E_M. The Fermi theory normally gives $f'(E')$, the distribution of electrons in energy, but since $E' + E = E_o$, f(E) may be obtained by simple substitution.

$$f(E)dE = k(E_o - E)\sqrt{(E_o - E)^2 - 1}\, E^2 dE \qquad \text{II-3}$$

$E_o = E_M + 1$, since the energies treated in β theory are total, rather than kinetic energies.

We shall rewrite Equation II-1 as

$$\sigma^{o}(E, \Delta) = K(E - \Delta)\sqrt{(E - \Delta)^2 - 1} \qquad \text{II-4}$$

Then

$$\sigma(E_M, \Delta) = kK \int_{1+\Delta}^{E_M} (E_o - E)\sqrt{(E_o - E)^2 - 1}\, E^2 (E - \Delta)\sqrt{(E - \Delta)^2 - 1}\, dE$$

This integral is too involved to solve exactly, and it is simple to show that if the ones are dropped from under the two radicals, the values of σ obtained in this way are within a few percent of their correct values. Certainly the one which appears in the radical with Δ may be dropped without appreciable error, as this only affects the form of the relationship between σ and E in the range of energies where σ^o is almost zero; therefore no appreciable contribution to the integral comes from this energy range. When the other 1 is dropped, a small quadratic "tail" is added to the neutrino spectrum at high energy, extending the upper limit by 1/2 Mev. The net effect of this approximation will be to increase $\sigma(E_M, \Delta)$ by a few per cent. In this approximation, we have

$$\sigma(E_M, \Delta) = kK \int_{\Delta}^{E_o} (E_o - E)^2 (E - \Delta)^2 E^2 dE$$

$$\sigma(E_M, \delta) = K(E_M + 1)^2 \left[0.285 - \delta + \delta^2 - \delta^5 + \delta^6 - 0.285\,\delta^7\right] \quad \text{II-5}$$

where $\delta = \Delta/(E_M + 1)$.

The function $\sigma(E_M, h)$ is shown in Figure 3. h is the height (above the ground state of A^{37}, of the level in A^{37} which is made by neutrino capture, in Mev. For the ground state of A^{37} (h = 0), $1 + \Delta_o$ is the measured atomic mass difference $A^{37} - Cl^{37}$. For comparison, σ^o is plotted in Figure 2. The constants which go into the evaluation of K are:

$Z = 18$

$\tau = 34.1$ days/.693 (expressed in seconds)

$\Delta_o = 0.65$ (H.T.Richards and R.V.Smith, Phys. Rev. 74, 1257 (1948)

To find the average cross section for pile neutrinos, we must know the "spectrum" of β-emitters in the fission process. And since σ rises quadrically with energy, we are most interested in the high energy beta emitters, i.e. those with short lives. Although data concerning such isotopes are hard to obtain, a

number of good experiments have been done, which shed a good deal of light on the shape of the high energy spectrum. Way and Wigner[x] have published a very complete

[x] K. Way and E. Wigner, Phys. Rev. 73, 1318 (1948)

analysis of all the pertinent experiments, and suggest the spectrum $N(E_M)$, shown in Figure 4. The same figure shows the function $N(E_M)\sigma(E_M)$, for various values of h. It may be seen that the largest contributions to the integral of $N\sigma dE$ come from energies in the 5 Mev range of upper limits. This region is rather well explored, so the value of the integrals, which are of course proportional to $\overline{\sigma}$, cannot be much in error. If the very high energy neutrinos were responsible for a large fraction of the average cross section, there would be a large possibility of error in the calculated value. The average cross section for pile neutrinos has been evaluated by numerical integration of the equation

$$\overline{\sigma}(h) = \int_0^\infty \sigma(E_M,h)N(E_M)dE_M \qquad \text{II-6}$$

This function is plotted in Figure 5. Its value when h = 0 will be used in all calculations, since the contributions from states with finite values of h will be shown to be small. (This statement cannot be made with absolute certainty, but it is the only reasonable assumption which can be made.) We will therefore use the value

$$\overline{\sigma} = 2.0 \times 10^{-45}\ \text{cm}^2 \qquad \text{II-7}$$

The true value of $\overline{\sigma}$ depends upon the level structure in A^{37}, and is given by

$$\overline{\sigma} = \overline{\sigma}(0) + \sum_{i=1}^{\infty} \overline{\sigma}(h_i)$$

where h_i is the excitation energy of the ith level in A^{37}. For all practical purposes, one need only take the sum over the levels which combine with the ground

state of Cl^{37} in allowed transitions. (The contributions from states giving forbidden transitions will be lower by a factor of about 100^n, where n is the degree of forbiddenness.) There is no known way to find the locations of such levels, but one may use as a guide the locations of levels in S^{33}, which differs from A^{37} by a single α-particle. According to a theory of Wigner's[x], nuclei in this part

[x] E. Wigner, Phys. Rev. 51, 106 (1937)

of the periodic table, which differ by one α-particle, should have about the same level structure within a few Mev of their ground states. In addition, Wigner calculates a distribution of levels, which has recently been found to be in good agreement with the experimental work on S^{33}. The levels in S^{33} have been mapped by Davison[xx], who observed the proton energies in the d,p reaction on S^{32}. He finds

[xx] P. W. Davison, Phys. Rev. 75, 757 (1949)

12 levels about equally spaced in the range from 0 to 6 Mev. There is only one level between 0 and 2 Mev, at about 0.8 Mev. If we assume the same level spacing in A^{37}, and make the reasonable assumption that the lowest level will not combine in an allowed transition with Cl^{37} and that 1/2 of the other levels will have similar properties, then the total neutrino cross section will be increased by about 25°/o over the value used in the main body of this paper. By a fortunate arrangement of levels, the total cross section might conceivably be increased by almost a factor of 2, but one should not count on more than a few per cent. Any such increase will be considered to balance the neglect of certain factors in the calculation of the A^{37} effect. For example, the fact that the Auger coefficient is 93°/o, rather than 100°/o has been ignored, and the end effects in the counter have not been considered. Although it is not correct to work to such a degree of accuracy in a proposal of this sort, one might as well balance small gains against small losses. Therefore any gain in cross section from an unexpectedly favorably

located excited state, will be considered as a bonus to be welcomed, but not counted on.

Let us now look at the question of the possible forbiddenness of the $A^{37} \rightarrow Cl^{37}$ transition. According to Konopinski, one decides whether a transition is allowed or forbidden by evaluating a quantity which is inversely proportional to the square of the matrix element $|H|^2$. This quantity is denoted by ft, where t is the half life for the transition, and f is a function of the energy released in the transition, and the atomic number of the element which undergoes decay. If all matrix elements had the same value, all values of ft should be the same. Since the maximum possible value of $|H|^2$ should be unity, there should be a certain minimum value of ft, corresponding to allowed transitions. If this simple interpretation were correct, one could conclude that almost the only allowed transitions were found in the so-called Wigner series of positron emitters. This is because these isotopes all have ft values of a few thousand, and the next smallest ft values are about ten times as large. This drop in the matrix elements for allowed transitions (for this is the logical way of explaining the second group of isotopes with higher values of ft) comes from the fact that the matrix element for an allowed transition involves the heavy particle states of the initial and final nuclei. If the initial and final states are almost identical, as is the case for the Wigner series, then the wave functions of the heavy particle in the two states "overlap" to a high degree and the matrix element for an allowed transition has its maximum possible value. But if the wave functions for the initial and final states of the nucleon are different, then the matrix elements will be decreased, even though the transition is allowed.

On the basis of this qualitative explanation, one would expect that the ft value for an allowed A^{37} transition would be at least 10 times as great as that for Be^7, an electron-capturing member of the Wigner series. If the A^{37} were for-

bidden, its ft value should be another factor of about 100 times greater, or at least 1,000 times the Be^7 ft. The experimental ft for A^{37} is 50 times that of Be^7. One therefore concludes that A^{37} is allowed, and that the calculated value of $\bar{\sigma}$ is correct.

The uncertainty in the value of $\bar{\sigma}$ if the transition is forbidden, comes from the assumption that the matrix elements for the forward and reverse reactions are equal. In the first order theory, the forbidden matrix element is zero; its observed finiteness comes from higher order terms which depend, for example, on the neutrino and electron wave lengths. Since all neutrinos have the same energy in the "forward reaction," one has experimental information about the matrix element at that one energy only. The neutrinos which contribute most to the backward reaction have a much higher energy and it is possible that their associated matrix elements could be higher. But the matrix element could not be more than 50 times larger, according to the argment regarding the ft values, and it is more likely that 5 times would be the upper limit.

APPENDIX III

Calculation of Neutrino Flux

$$10^8 \text{ watts} = 10^{15} \text{ ergs/second}$$

$$= \frac{10^{15}}{1.6 \times 10^{-12}} = 6.25 \times 10^{26} \text{ electron volts/second}$$

$$= \frac{6.25 \times 10^{26}}{2 \times 10^8} = 3.12 \times 10^{18} \text{ fissions/second}$$

Way and Wigner take the average number of beta decays per fission to be 6.3. Therefore 10^8 watts corresponds to the emission of $3.12 \times 10^{18} \times 6.3 = 1.97 \times 10^{19}$ neutrinos per second.

The neutrino flux is then

$$nv = \frac{1.97 \times 10^{19} P}{4\pi R^2}$$

$$\boxed{nv = \frac{1.70 \times 10^{15} P}{D^2}}$$

P = power in 10^8 watts

R = distance in cm.

D = distance in feet

The neutrino flux from the sun is of the order of 10^{10}. But the sun neutrinos have energies much lower than those of the average pile neutrinos, so their effects will be quite negligible.

APPENDIX IV

Cosmic Ray Induced Background

Argon^{37} can be made from Cl^{37} in p,n reactions, by any protons associated with the cosmic radiation. The most obvious sources of such protons are cosmic ray stars. Such stars, or nuclear explosions, have been observed in cloud chambers, ionization chambers, and photographic plates. The data to be used in the calculations of background effects come, for the most part, from experiments with photographic emulsion plates. Several men have determined the rate of production of stars, as a function of elevation, and the latest measurements are probably reliable. The earlier experimenters did not appreciate the importance of the fading of the latent image, and in an effort to increase the density of stars in a given plate, exposed their emulsions for times long compared to the "fading time." For this reason, their estimates of the star production rate are too low.

Most of the men who have determined the star production rate in emulsion have also measured the average flux of single proton tracks in the same set of plates. There are two reasons for believing that practically all the single protons come from stars , which may be in the emulsion, the glass backing, or the air. In the first place, the absorption coefficient of the "star producing radiation" is identical to that of the "proton producing radiation." Secondly, the flux of single tracks is what one would predict from the density of stars, the average range of the observed star protons, and the average number of protons emitted per star.

Until very recently, there was no good evidence as to the nature of the star producing radiation. In his book on cosmic rays, Heisenberg identified it as the soft component, and more recently, Perkins has believed that it was a new

type of unstable particle which decayed in flight. Perkins[x] measured the in-

[x] D. H. Perkins, Nature, 160, 707 (1947)

tensity of stars as a function of altitude, and found that in the atmosphere, the absorption coefficient had a constant value. Expressed as a mean free path, it was 150 gm/cm^2. He then piled lead over some photographic plates exposed at sea level, to extend his absorption measurement to greater effective depths from the top of the atmosphere. When he plotted his results on semi-log paper, he had a straight line decrease in intensity from 5.7 meters of water equivalent from the top of the atmosphere, to 10 meters (sea level). From 10 meters to 13 meters (H_2O equivalent for the Pb), he had a very small change in intensity, and the probable errors on his individual points were such that one could not exclude the possibility that the 300 gms/cm^2 of Pb had not changed the star intensity at all. Perkins therefore concluded that the stars might result from the decay of an unstable particle in flight, and that distance was more important than mass, in reducing the intensity of the star producing radiation.

If this conclusion had been correct, the proposed experiment would be impossible with presently available piles, all of which are above sea level, and none of which produces a neutrino flux intense enough to "override" the cosmic ray background.

Fortunately, Perkins and his collaborators[x] have recently re-examined the

[x] J. B. Harding, S. Lattimore, T. T. Li, and D. H. Perkins, Nature 163, 319(1949)

problem and have found the following results. The star producing radiation has the following mean free paths in air, ice, and lead:

$$\lambda_{air} = 150 \pm 10 \text{ gm/cm}^2$$

$$\lambda_{ice} = 200 \text{ gm/cm}^2$$

$$\lambda_{Pb} = 315 \pm 5 \text{ gm/cm}^2$$

The experiments with ice were done on the Jungfraujoch, and log I was a linear function of thickness over a factor of 15 in intensity. This result shows that the stars are not due to an unstable radiation, and suggests that the absorption is due entirely to nuclear collisions - presumably those collisions which produce the stars. The calculated oxygen cross section is $\sigma_o = 0.15$ barns. This is about what one would expect for high energy protons or neutrons, and it is very probable that the star producing radiation is the "tail" of the primary cosmic ray protons. Although Perkins does not so identify it, it may be shown that the known flux of primary protons would give the observed intensity of stars as a function of absorber thickness, if the star producing cross sections were close to 0.15 barns. For the purposes of this discussion, the most important result is that the star producing radiation may be attenuated by passage through matter.

To calculate the cosmic ray induced A^{37} background, in an unshielded tank of CCl_4, we will use the following data from the Perkins group:

$$S = 1.0 \text{ stars/cc day in emulsion at sea level.}$$

$$(nv)_{obs} = 0.3 \text{ protons/cm}^2 \text{ day (energies below 50 Mev).}$$

The energy distribution of protons from stars, as determined by Perkins[x]

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[x] Perkins, Nature 160, 299 (1947)

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shows a peak at 10 Mev, and an average energy of about 15 Mev. However, the average range corresponds to a much higher energy, since $R = kE^{1.75}$. Even though the fraction of the protons with energies above 20 Mev is very small, the contribution of these protons to the mean range is very substantial. By

numerical integration of Perkins' curve times the range-energy function in emulsion, the average range R is found to be 0.32 cm.

One should be able to correlate the values of S, R, nv, and P, the average number of prongs per star. To be sure that Perkins' data were being interpreted correctly, this had to be done, as on first sight, the values of S and nv did not seem to be in accord. One might think that nv should be equal to SRP, and this is true if nv is defined in the manner familiar to pile workers. But Perkins' value of $(nv)_{obs}$ may be shown to be smaller than the common definition of nv by a factor of two. In addition, his quoted value of $(nv)_{obs}$ does not include protons over 50 Mev, while the value of R obtained from his data, does include such protons. When these correction factors are applied, one gets a consistent set of numbers relating to star protons in emulsion. When they are changed to apply to the case of CCl_4, we have

$$S = 0.8 \text{ stars/cc day}$$

$$P = 4.0$$

$$R = 0.5$$

$$nv = PRS = 1.6/\text{cm}^2 \text{ day}$$

$$\sigma = 10^{-25} \text{ cm}^2 \text{ (for the p,n reaction)}$$

The cross section is an "educated guess," but it is about the best one can do with the available data. The activity of A^{37} after a bombardment of 2 half lives will then be

$$A_2 = \frac{3}{4} SRP\sigma \frac{N_o V \rho}{M}$$

where N_o is Avagadro's number, M is the molecular weight of CCl_4, V is the volume of the tank, and ρ is the density of CCl_4. (The abundance of Cl^{37} just cancels the factor of 4 which would come from the number of Cl atoms in a molecule.) On substituting the numbers, we find

$$A_2 = 2 \times 10^4 \text{ counts per day}$$

This is probably an upper limit, as it is known that the value of σ decreases as the proton energy increases, because of competing reactions. This is a most important fact, as the photographic plates used in Perkins' work do not show protons with energies above 100 Mev. The flux of primary protons (if they are really the same as the star producing radiation) can be shown to be several hundred times as great as that of the observed star protons. If they had the same p,n cross section, the value of A_2 would be greater than that listed above, by the same ratio. But recent work by the Berkeley Chemistry Group has shown that at 350 Mev proton energy, the p,n cross sections are of the order of 10^{-27} cm^2, or less. They have not been observed definitely, so one can merely set upper limits. This is sufficient, however, to indicate that the insensitivity of the photographic emulsions to high energy protons does not deprive us of essential information as to the background production of A^{37}. The higher energy protons produce stars (spallation reactions), and only very rearely strike a nucleus with such a "glancing blow," that a single neutron is ejected. In the light of this new information, it is probably more correct to use 10^4 as the background activity.

Since 10^4 counts per day is very large compared to the expected neutrino induced counting rate, the problem of shielding is of the greatest importance. If we want the background to be 1 per day, we must place $\ln 10^4 = 9$ mean free paths of absorbing material over the CCl_4 tank. If the shielding were water, its thickness would be 18 meters, or 60 feet. This is clearly an impractical type of shield to build expressly for one experiment. Another method of shielding immediately comes to mind; the pile itself could be used, by burying the CCl_4 underground. No numbers are available on the size of the Hanford piles, but published photographs of the Argonne and Harwell piles indicate that the exterior dimensions of the shields are approximately 40 feet. As will be seen

in Appendix VI, which treats the background effects due to fast neutrons, the CCl_4 tank must be at least fifteen feet from the edge of the active volume of the pile. Since the density of graphite is more than 1.5, it is apparent that the pile will give adequate shielding against vertically directed star producing radiation. If one draws a diagram of a tank placed 15 feet below the center of the pile, he finds that star producing radiation may strike the tank from zenith angles greater than 45°, without passing through the pile. However, there is still a large thickness of earth in the direct path of the cosmic radiation, and the longer path of the rays in the atmosphere gives an additional attenutation which makes up for the smaller path in solid material. Until the exact dimensions of the piles are known, it is impossible to evaluate the background, but the attentuation should be more than adequate.

In the discussion above, it has been tacitly assumed that there is only one kind of proton-producing radiation, with a single absorption length. It is well known that cosmic rays may be observed at depths under the earth equivalent to many hundreds of meters of water. If this radiation were capable of producing A^{37}, the shielding problem would be hopeless. It is fairly well established that the very penetrating component consists of high energy μ-mesons. When these mesons decay in flight, they turn into high speed electrons, which then produce showers. Such showers can produce no background directly, since neither electrons, nor gamma rays, can make A^{37} from Cl^{37}. Positive mesons could theoretically be absorbed by Cl^{37} nuclei, to give A^{37}. This is of no practical importance, since the interaction of μ-mesons and nuclei is so weak. The positive μ-mesons have a vanishingly small chance of inducing the reaction when traveling at high speed, and after they are brought to rest, they decay; their positive charge keeps them out of the nucleus.

One additional mechanism by which the penetrating component of cosmic rays could produce A^{37} must be investigated. The showers observed underground are in equilibrium with the high energy μ-meson component, and contain high energy gamma rays which could liberate protons by photo-disintegration processes. Since there is no effective shield against this component of the cosmic radiation, its effect will have to be small at sea level, if the proposed experiment is to be successful. Unfortunately, the cross sections for γ,p reactions at high γ-ray energies are not well known, and the flux of γ-rays underground is not well known either. The following method of evaluating the proton component under 20 meters of water equivalent should at least give the proper order of magnitude. The μ-mesons can produce fast electrons by three distinct processes: (1) decay in flight, (2) radiative collisions followed by pair production, and (3) "knock-on." The third process produces low energy electrons, compared to the binding energies of protons, and will therefore be disregarded.

The radiative processes will be considered first. Since the meson has a mass about 200 times that of the electron, it will radiate $\overline{200}^2$ times less than an electron. An electron undergoes a radiative process on the average in a distance equal to "one shower unit," so a meson will go 40,000 shower units before radiating. The shower it makes will extend over an average length of about 4 shower units. (All discussion is in terms of mesons with the most probable sea level energy of about 10^9 ev.) The average number of γ-rays in the shower will be about 10, so the flux of γ-rays relative to mesons will be $4 \times 10/40{,}000 = 10^{-3}$. The flux of protons relative to that of γ-rays will be in the ratio of $(\sigma_{\gamma,p} \text{ to } \sigma_{pair})_{Cl}$, or approximately $10^{-28}/2 \times 10^{-24} = 5 \times 10^{-5}$. (The γ,p cross section is only an estimate based on recent synchrotron and betatron work, but the ratio of the two cross sections checks approximately with

the ratio of proton to electron tracks, as observed in cloud chamber pictures of showers[x], when corrected for the theoretical ratio of γ-ray quanta to

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[x] W. Fretter, private communication.

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electrons in showers. Since this is really the important number for evaluating the background, we can have some confidence in the method of analysis.) The ratio of photo-proton to meson fluxes is then 5×10^{-8}. From Rossi's review article on cosmic rays[x], we have the meson flux at 20 meters of H_2O

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[x] B. Rossi, Rev. Mod. Phys. 20, 544 (1948)

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equivalent below sea level. This flux is 1.3×10^{-3} per cm^2 second steradian. The total flux is then approximately 5×10^{-3} per cm^2 second, or 4.3×10^2 per cm^2 day. The photo-proton flux will therefore be $4.3 \times 10^2 \times 5 \times 10^{-8} = 2.2 \times 10^{-5}$ per cm^2 day.

In the section on "star background," we found that a proton flux of 1.6 per cm^2 day gave rise to a background activity of 10^4 counts per day, so the radiative processes of mesons will give rise to a background of $10^4 \times 2.2 \times 10^{-5}/1.6 = 0.14$ counts per day. It is obvious that the numbers in this section are considerably more crude than those pertaining to the star background, but it is highly unlikely that they are off in an unfavorable direction by a factor which would make the experiment impossible.

The second important process by which mesons generate showers is by decay into electrons during flight. The mean life for this process is 2×10^{-6} second in the moving system. An observer in the laboratory finds this time to be increased by the ratio E/uc^2, which is 10 for a 10^9 ev meson. The average distance a meson goes before decaying is then $10 \times 2 \times 10^{-6} \times 3 \times 10^{10} = 6 \times 10^5$ cm. One shower unit in CCl_4 is about 25 cm, so the "decay distance"

is 2.5×10^4 shower units. Since the "radiative distance" is 4×10^4 shower units, the background due to the decay process should be about 4/2.5 times that due to radiation. Actually, this is an under-estimate, since the decay particles will give larger showers than the bremstrahlen. But since the γ,p cross sections decrease with increasing energy, this should not be a large effect.

This section may be concluded by saying that according to the most realistic estimate of the cosmic ray effects, the A^{37} background due to these various processes should not be much greater than one count per day.

APPENDIX V

Background from Radioactive Impurities

α-particles are observed to be emitted from surfaces of any material used in the construction of ionization chambers. They are attributed to heavy atom impurities, and the ranges of the alpha particles are found to be identical to those of the known, naturally occurring radioactive series. The percentage of radium and thorium which is present in a material may be determined by counting the number of α's per cm^2 of surface per day. Typical values of these percentages will be listed later in this appendix.

The α-particle background in the CCl_4 will not give rise to A^{37} in a direct process. The reaction $Cl^{35} + \alpha \rightarrow A^{37} + d$ is endothermic, with a threshold energy in the neighborhood of 10 Mev. This type of reaction has never been observed, but that would not be a sufficient reason for neglecting it, if it were energetically possible. Since it is excluded on energetic grounds, one may then look for secondary reactions, which are initiated by α-particles.

Protons may arise from the reactions $Cl^{35,37} + \alpha \rightarrow A^{38,40} + p$. Protons have been observed from α-particle bombarded Cl, by Rutherford and later workers. The yield is not a rapid function of α-particle range, using the α's from naturally radioactive substances. The best estimate of the proton yield is about 10^{-6}. One may neglect carbon as a contributor to the proton flux, as the α,p reaction on carbon is not observed when radioactive α-particles are used.

The protons may make A^{37} by a p,n reaction on Cl^{37}. The yield on pure Cl^{37} is probably somewhat greater than 10^{-4}, but not so large as 10^{-3}. The overall yield will not be underestimated if the p,n yield on normal Cl is taken

as 10^{-4}. The overall yield of A^{37} from α-particle bombardment will then be taken as $10^{-6} \times 10^{-4} = 10^{-10}$.

We may now calculate the maximum amount of α-contamination which is permitted in the CCl_4, if the α-induced A^{37} background is to be kept below 1 count per day. This will obviously be 10^{10} α-particles per day in 4×10^7 gms of CCl_4, or 2.5×10^2 α-particles per gram day of CCl_4. Since all A^{37} activities are calculated for a 68 day bombardment, we may multiply 2.5×10^2 by 4/3, to give 3.3×10^2 α's per gram day, or 4×10^{-3} α's per gram second. To convert this into more familiar units, we will assume that the α's come from radium and its decay products, and calculate the radium impurity. Assuming 5 α's per disintegration of radium (Ra + 4 daughter substances) we can tolerate

$$\frac{4 \times 10^{-3}}{5 \times 3.7 \times 10^{10}} = 2 \times 10^{-14} \text{ gms Ra/gm } CCl_4$$

Normal samples of copper and iron contain an average of 10^{-14} gms Ra/gm metal. There are reasons to believe that the radium content of CCl_4 will be much less than that of ordinary metals, but even without those reasons, the A^{37} background would not be serious. The chlorine which goes into the manufacture of CCl_4 is probably derived from sea water, and the radium content of sea water is about 1% of that of ordinary materials. Another important consideration is that CCl_4 should be easily purified from radium, by distillation, whereas chemical methods of purification usually introduce as much impurity as they eliminate. Although the α-particle effect is not negligible, it should not contribute appreciably to the difficulties of the experiment.

For the sake of completeness, one should investigate other radioactive sources of protons. Both β-rays and γ-rays are capable of releasing protons from stable isotopes, but they must have energies greater than the binding

energies of the protons they release. Since this condition is not satisfied in the case of β- and γ-rays from the naturally radioactive series, one may neglect such effects in calculating the background.

APPENDIX VI

Background from the Pile

One is in the habit of thinking that the fast neutron flux outside an operating pile is essentially zero. But in connection with an experiment where one must worry about secondary reactions from α-particle impurities, it is certainly not safe to assume that a neutron flux is zero just because it is ordinarily unobservable. Neutrons cannot by themselves produce A^{37}, since they have no charge. But protons from n,p reactions can give the now-familiar p,n reaction on Cl^{37}.

Instead of calculating the activity of A^{37} due to neutrons from the pile, it is more instructive to proceed as in the last appendix, and calculate the maximum neutron flux which can be tolerated. If this turns out to be greater than the actual flux, one may then estimate the additional shielding required to reduce the neutron flux to the allowed value. Since the "half thickness" for attenuating fast neutrons from a pile is about 4" of concrete, one can easily provide for a large attenuation without increasing D (the distance from pile center to CCl_4 tank) in a drastic manner.

The reaction $Cl^{35}(n,p)S^{35}$ is exothermic, so neutrons of any energy are capable of releasing protons of slightly greater energy, in the CCl_4 tank. But since the reaction $Cl^{37}(p,n)A^{37}$ has a threshold of about 1.7 Mev, neutrons below 1 Mev cannot contribute to the production of A^{37}. The fraction of the incident neutrons which give rise to p,n reactions in A^{37} is not known with certainty, but it is certainly not greater than 10°/o. (This seems a safe upper limit, in view of the elimination of neutrons below 1 Mev, and because of the absorption and degredation of the flux in passing across the 8-foot diameter of the tank.) If we assume a p,n yield of 10^{-4}, from the last

appendix, the overall yield of A^{37} from neutrons is 10^{-5}. Since we want not more than one A^{37} decay per day, we can tolerate no more than 1.2 fast neutrons per second across the projected area of the tank. For a tank 16 feet long and 8.4 feet in diameter, holding 40 metric tons of CCl_4, the projected area is 1.25×10^5 cm^2. The maximum allowable fast neutron flux is then 10^{-5} per cm^2/second. For health protection reasons, the flux outside the pile will be of the order of 10 per cm^2 second, or less. To reduce the flux to 10^{-6} of this value requires the addition of about 20 "half thickness" of absorbing material. Since the half thickness is approximately 4" of concrete, or at most 8" of dirt, the additional shield should be less than 13 feet thick. This is a very conservative estimate, and gives a not-unreasonable additional shield.

It is worthwhile at this point to look for other sources of A^{37} background. As has been stressed earlier, the only particles capable of producing A^{37} directly from chlorine are those carrying at least one positive charge. The equivalent process of knocking out a negatively charged particle is not considered, since that particle would have to be (in the present state of our knowledge), a negative π meson. We must then inquire as to the possibility that protons are produced by other radiation from the pile. The only other process which comes to mind is the γ-p reaction. In the first place, γ,p cross sections are much smaller than n,p cross sections. Secondly, the absorption coefficients of γ-rays from the pile are greater than those of neutrons, so a shield sufficient to reduce the neutron intensity to a negligible intensity will make the γ-ray effects still more unimportant.

The production of A^{37} from impurities in the CCl_4 should be investigated. One may neglect argon as an impurity, since the whole basis of the experi-

ment is that almost every atom of argon in the tank may be removed by the boiling and "sweeping" to be discussed in Appendix VIII. Therefore, if there is any argon in the tank, for neutrons to interact with, it will not mean that the background is increased, but rather that there will be no effect to observe in the first place.

Fast neutrons may produce the reaction $Ca^{40}(n,\alpha)A^{37}$. Since we have postulated a neutron shield thick enough to reduce the n,p reactions to the point that the secondary p,n reactions on Cl^{37} cause no increase in background, it is easy to show that the n,α reaction on Ca^{40} will be of no consequence. The n,p yield was taken as 10°/o, so there would have to be about 10°/o Ca^{40} in the tank, if it were to be of importance in this respect.

APPENDIX VII

Reduction of Counter Background

In the last three appendices, the reduction of the A^{37} background has been considered. A more usual type of background is that introduced by the counter itself. The reader has probably been surprised to see that a single counter background, in an electron counting experiment, has been set at one per day. Libby, in his recent work on naturally occurring radio-carbon, has succeeded after a great deal of work, in reducing his single counter background from 400 per minute to 7.5 per minute. He uses two tons of shielding material around his counter, and employs anti-coincidence counters to eliminate cosmic ray background. He has probably done the most thorough job in cutting down a single background, but his result falls short of the one per day value by a factor of 10^4.

The counter to be used in the proposed experiment differs from Libby's in two important respects. Since the volume of argon to be taken from the tank is essentially zero (a few thousand atoms at most) the counter volume may be made arbitrarily small. The counter is assumed to have a diameter of 3 millimeters and a length of 1 cm. Its wall area is therefore 1 cm^2, or 400 times less than Libby's. So, on a relative basis, the background of the small counter must be reduced by a factor of only 25. Several things make this possible. The electrons to be counted in the A^{37} experiment have a unique energy of 2.8 Kev, so they could not penetrate the thinnest counter walls. (The "L-capture" in A^{37}, which has recently been observed by Pontecorvo, gives a few very low energy electrons, but this has no effect on the conclusions reached in this appendix.) This means that the counter walls can be made very thin (a few mils of solid material), which gives two immediate

benefits. In the first place, the radioactive contamination in the walls is reduced in the ratio of the wall thickness. In the second place, the contamination β-rays which originate in the walls and give counts in the small counter, are now free to pass through the walls and enter the surrounding anti-coincidence counter. Since Libby is counting the β-rays from radio-carbon, he has to keep the wall thickness between his active counting volume and his shield counters, greater than the C^{14} maximum range. This necessity probably accounts for his background that is not eliminated by the anti-coincidence shield.

The proposed counter will be entirely surrounded by a multi-wire proportional counter. This shield counter will respond to all ionizing radiation which activates the small counter and has enough range to pass through the small counter wall. In this class of radiation, we may include a large fraction of the β-rays from the small counter's wall, and all β-rays from outside the small counter (primary β's from the shield counter walls and gas, and secondary electrons from any γ-rays). The shield counter will also eliminate cosmic ray particles if the "gates" in the anti-coincidence circuits are long enough (10 μseconds) to take care of the decay electrons from μ mesons. As an additional, and probably unnecessary precaution, the whole counter setup could be placed under ground. (There is a deep tunnel near the Radiation Laboratory which has been used for a number of cosmic ray experiments; there is a good deal of space available in the ventillation ducts.)

α-particles from the counter walls do not belong to the class of particles which may be eliminated by the anti-coindicence shield counter. But since the small counter is to be used as a proportional counter, the α-particles may be eliminated by virtue of the large number of ions they make in the counter. If the counter is filled to one atmosphere of helium, the following

numbers are pertinent: an alpha particle makes 10,000 ion pairs, an A^{37} Auger electron makes 88 I.P., and a fast electron makes 3 I.P. The effective range of the Auger electrons is only 0.15 mm; since the diameter of the counter is 3 mm, there will be little "wall effect," and almost all of the Auger electrons will be counted. Since an Auger electron cannot make more than about 120 I.P. a discriminator circuit will be arranged to eliminate all ionization pulses corresponding to more primary ions. This will eliminate those "heavy particles" which do not penetrate the counter wall and thereby activate the anti-coincidence circuit. Pontecorvo has recently published several letters to the editor of the Physical Review, which show that the A^{37} Auger electrons may be counted quantitatively, and their individual energies measured, in a proportional counter.

So far, this discussion has been more or less qualitative. It will now be shown that the background from the counter walls is small enough so that the anti-coincidence arrangement might almost be eliminated. Since the latter is necessary to take care of cosmic rays, and β and γ-rays from the surrounding materials, it reduces the counter wall background from a small value to a negligible one. We will now calculate the counter wall background using the commonly accepted values of radioactive impurities. The standard value for the rate of emission of α-particles from copper and iron surfaces is one α per cm^2 day. (Recent work at the San Francisco Navy Radiological Laboratory has shown that electrolytic nickel has an α-counting rate of one-tenth this value.) Since the wall area of the small counter is one cm^2, the uncorrected α background will be one per day. The discriminator circuit should eliminate all but perhaps 2% of these counts, so the net α-particle background should be 0.02 counts per day.

The number of β-rays and conversion electrons emitted by the equilibrium decay products of radium is about equal to the number of α's. If the counter

wall has a thickness equal to the range of the α's, there will be twice as many β's per second leaving the surface as α's. (The β range is much greater than the thickness.) If one assumes that the counter wall is three α-particle ranges thick, which will be quite strong mechanically, the number of β-ray counts from impurities in the walls will be 3 x 2 x 1 = 6 counts per day. It is quite certain that the fraction of these beta rays stopped in the thin walls of the counter is less than 1/6, so the net counting rate with the anti-coincidence circuit operating will be certainly less than one per day.

APPENDIX VIII

Separation of A^{37} from CCl_4

In order for the experiment to be successfully performed, it is necessary to solve the separation problem. Several thousand atoms, in the most favorable case, will have to be separated quantitatively from 40 tons of CCl_4, and the argon atoms will then have to be introduced without loss into a very small proportional counter. Although this sounds like a most formidable and unprecedented operation, it does not differ greatly in magnitude from work which has been done in the past by experimental physicists. Paneth[x] irradiated 4 liters of a boron ester with slow neutrons, and separated the helium formed by the capture of slow neutrons. He was able to separate and make quantitative measurements on the helium, which was present in the amount of 2×10^{-8} cc, and 1.4×10^{-7} cc, in two of his experiments.

[x] F. A. Paneth, E. Glückauf and H. Loleit, Proc. Roy. Soc., A, 157, 412 (1936)

A similar experiment, with modifications suggested by E. Fermi, was performed during the war years under the author's supervision[x]. The separation of 4×10^{-4} cc of helium was effected, from about a liter of boric acid solution. Quite accurate absolute measurements of the neutron strength of Ra-Be sources were made in this way, so the author is familiar with the ease of separating small quantities of noble gases from solution. In addition, he has had recent experience in the rapid separation of N^{17} from deuteron-bombarded fluoride solutions[x]. In the N^{17} experiments, it was found that tracer amounts

[x] F. G. P. Seidl and S. P. Harris, Rev. Sci. Inst. 18, 897 (1937)

[x] L. W. Alvarez, Phys. Rev., April 15, 1949 (in press).

of this gas could be swept out of a water solution of NH_4F, without boiling the solution. Helium was bubbled through the solution, and the N^{17} was found in the gas stream, in very high intensity, although the half life of the N^{17} is only 4.2 seconds. This is not in the nature of a quantitative statement, but it does give an idea of the simplicity of extracting an inert gaseous product from a large volume of liquid. No significant increase in the activity was found when the solution was boiled, and if this is true in the case of CCl_4, a great simplification of the "chemical engineering" would result. Of cource, all such points may be tested in the laboratory, before the large scale process is designed.

The 4×10^7 grams of CCl_4 occupy a volume of about 2.7×10^4 liters, so the separation should not take more than 10^4 times as long as that required in the N^{17} case, if the CCl_4 were handled on a batch process. The separation will no doubt be simpler than this figure would indicate. It is very likely that if the CCl_4 were kept boiling for an hour or two, and if helium gas were bubbled through it at the same time, that more than 99°/o of the argon would be removed from the CCl_4. CCl_4 vapor may be removed from He and A by condensation, and A and He may be separated by passage through liquid air-cooled active charcoal. After the A^{37} has been trapped in the active charcoal, the latter may be warmed, and the A^{37}, together with the counter filling gas, may be transferred to the counter by means of a Toeppler pump.

Although it is not possible at the moment to design in detail the CCl_4 tank and the associated gas handling equipment, there is a good chance that the tank should be thermally insulated from the ground in which it is buried. Heat to boil the CCl_4 could be supplied electrically, and it should not be difficult to keep the liquid boiling all the time, in the unlikely event that it turned out to be advantageous.

It is the author's opinion that the separation problem will turn out to be relatively simple, even on the scale proposed. One might object that the efficiency of separation of tracer amounts of A^{37} from ton lots of CCl_4 might be lower than that achieved on the laboratory scale. If such objections were valid, the proposed experiment would not be a crucial test of the neutrino theory, since the collected activity might be low, while the produced activity was of the theoretically predicted magnitude. Fortunately, this point is easy to check. One can prepare samples of A^{37} by bombarding small amounts of CCl_4 with protons. Such experiments have been made using the 32 Mev protons from the Berkeley linear accelerator. The CCl_4, which was sealed in a glass ampule, was bombarded through the glass wall, and the A^{37} was boiled out of the CCl_4 and introduced into a counter. One could prepare A^{37} in a range of volumes of CCl_4, by bombardment with identical numbers of protons, and show that the collected activity was the same for all volumes. Finally, a small bombarded ampule of CCl_4 could be introduced into the large tank and cracked open under the surface of the liquid. The recovery technique could then be developed to the point where the known activity was collected from the large volume.

Acknowledgments

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LWA/vt

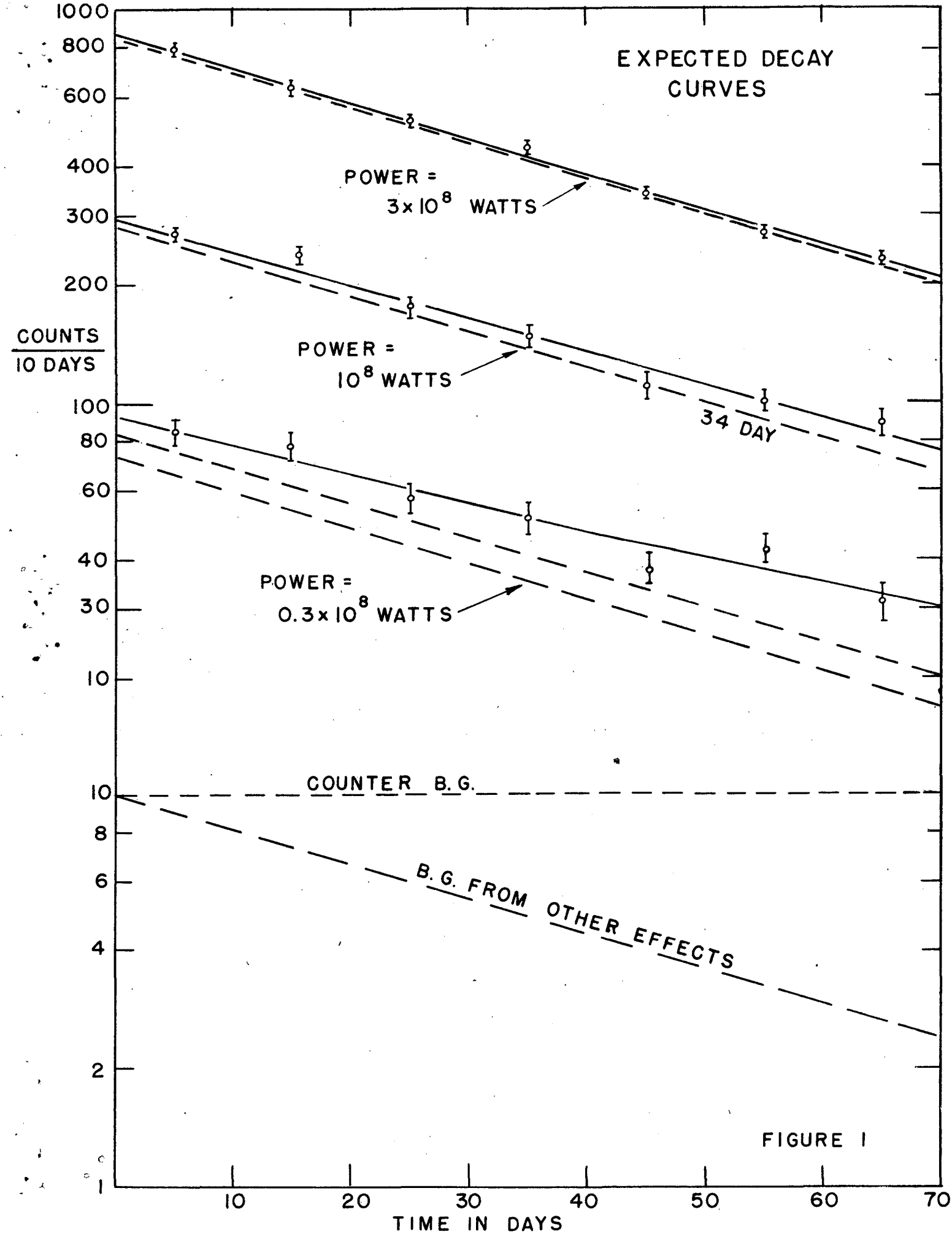

FIGURE 1

13610-1

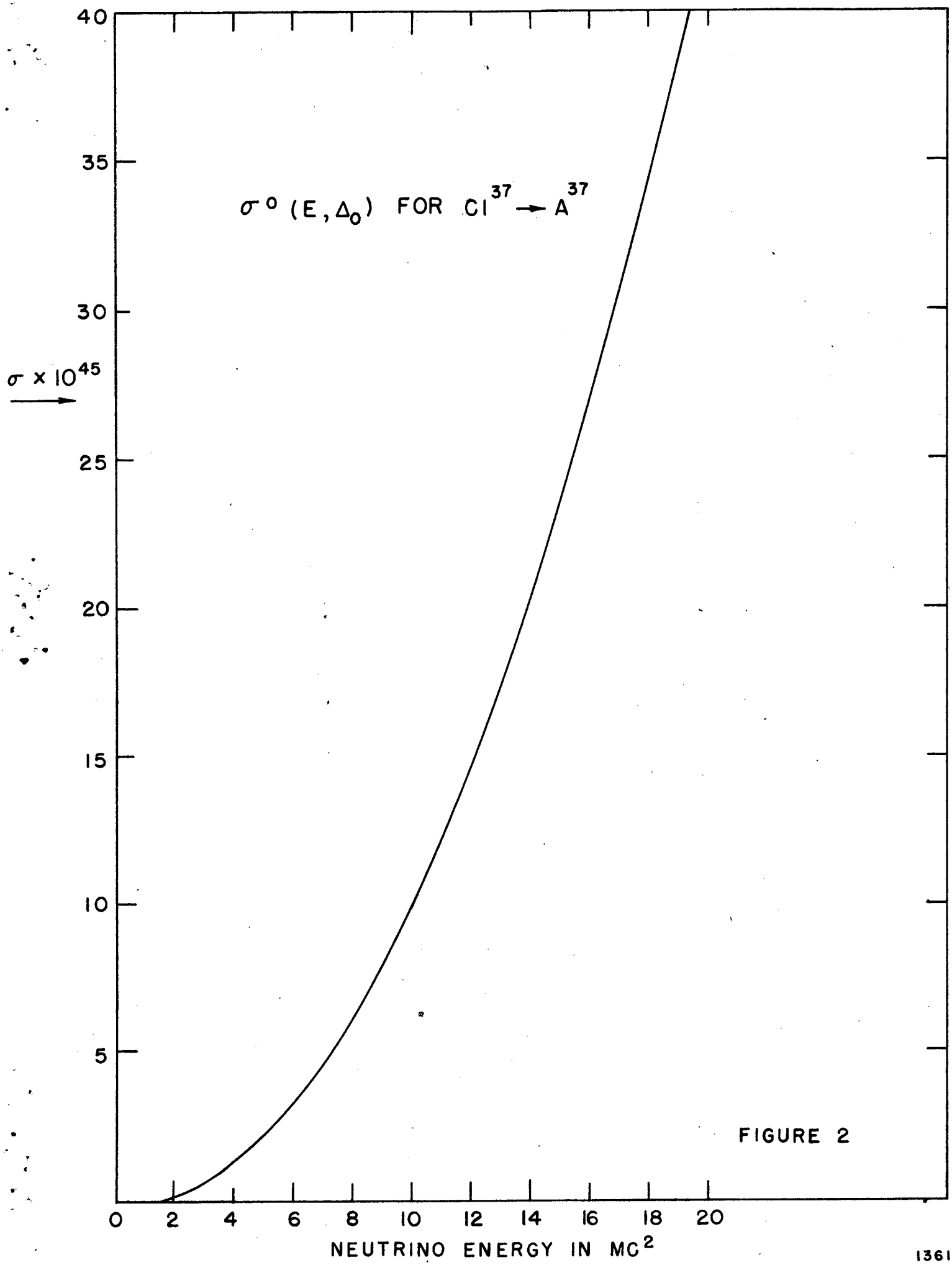

FIGURE 2

13611-1

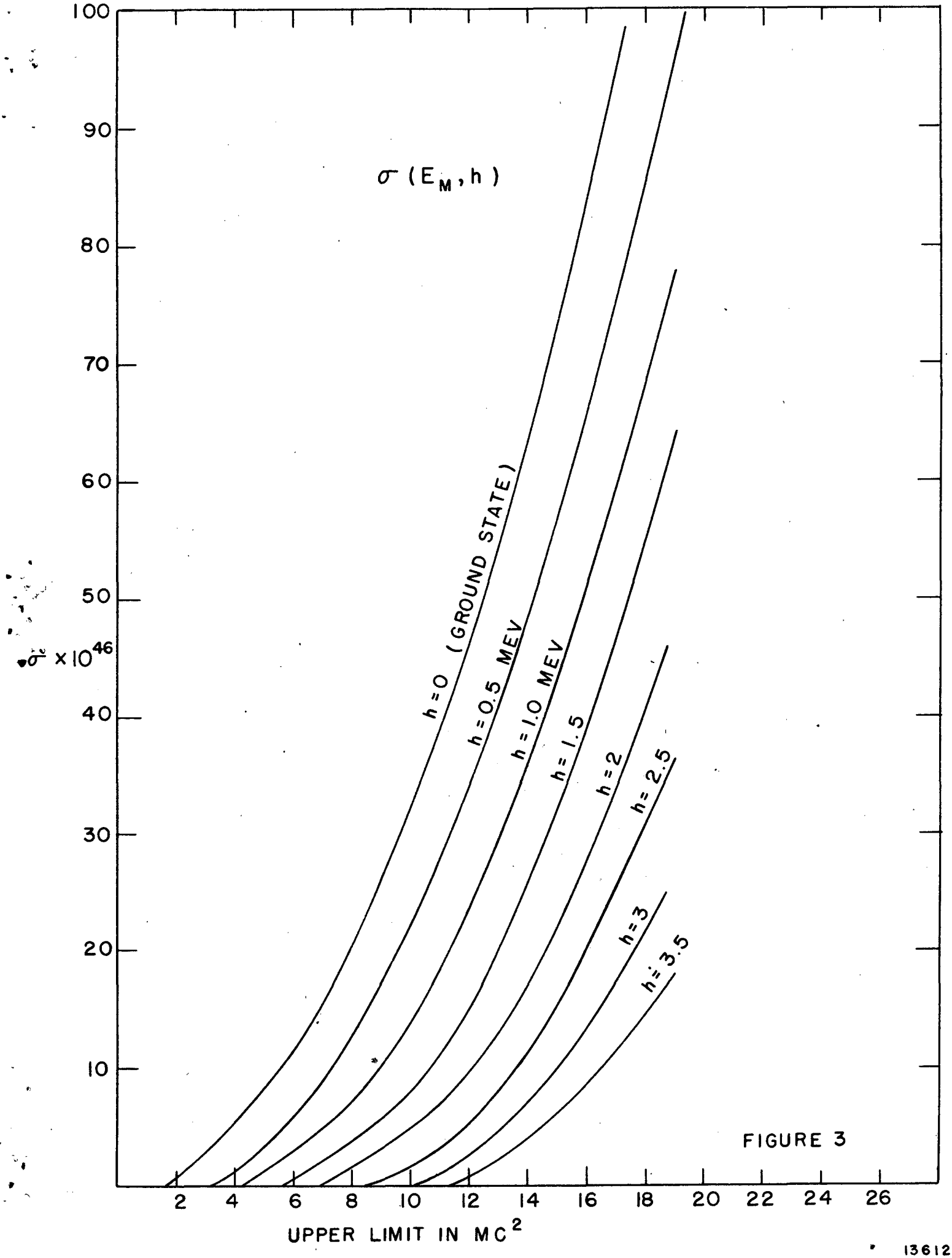

FIGURE 3

13612-1

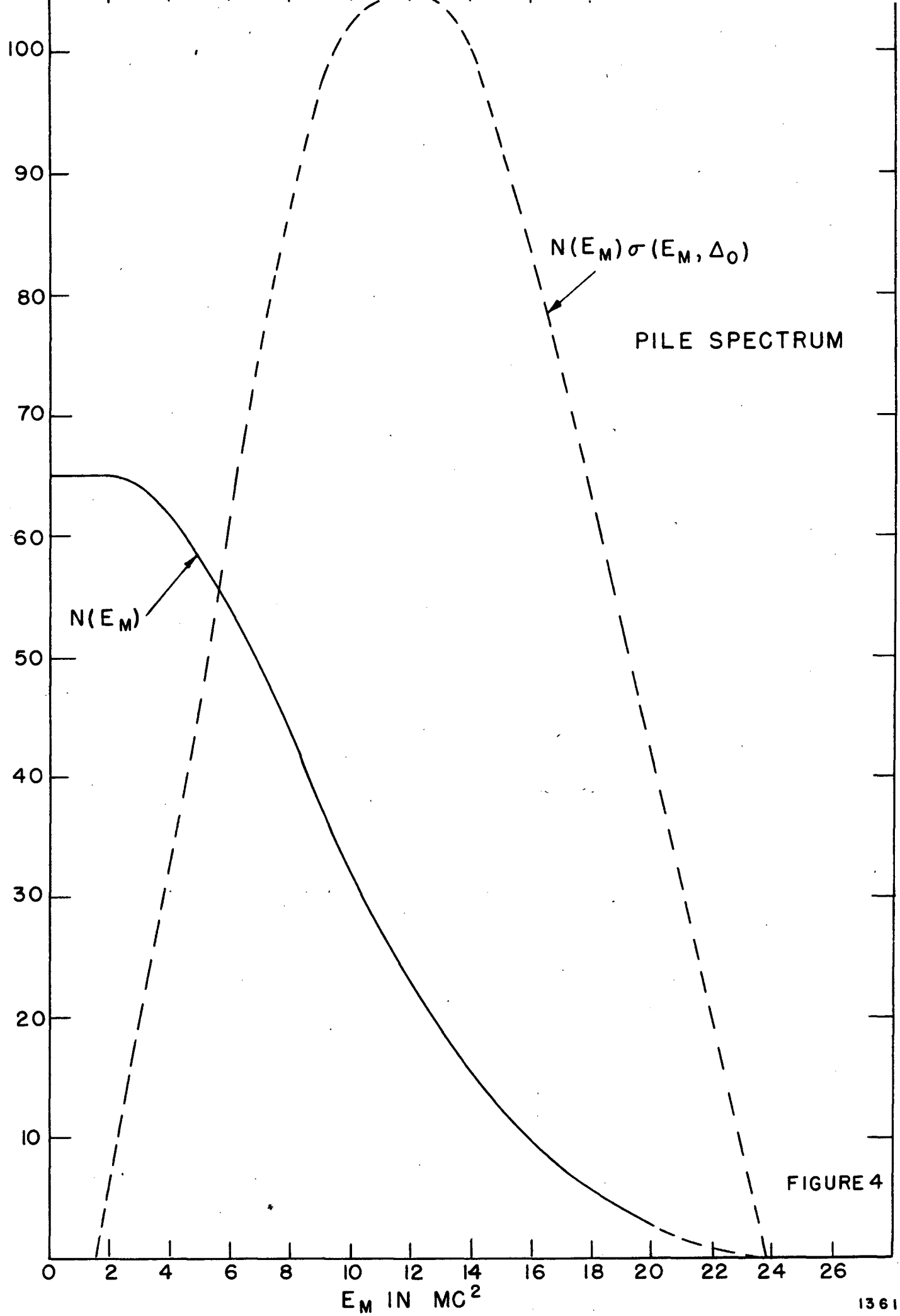
100
90
80
70
60
50
40
30
20
10
0
2
4
6
8
10
12
14
16
18
20
22
24
26
E_M IN MC^2
$N(E_M)\sigma(E_M, \Delta_0)$
PILE SPECTRUM
$N(E_M)$
13613-1

FIGURE 4

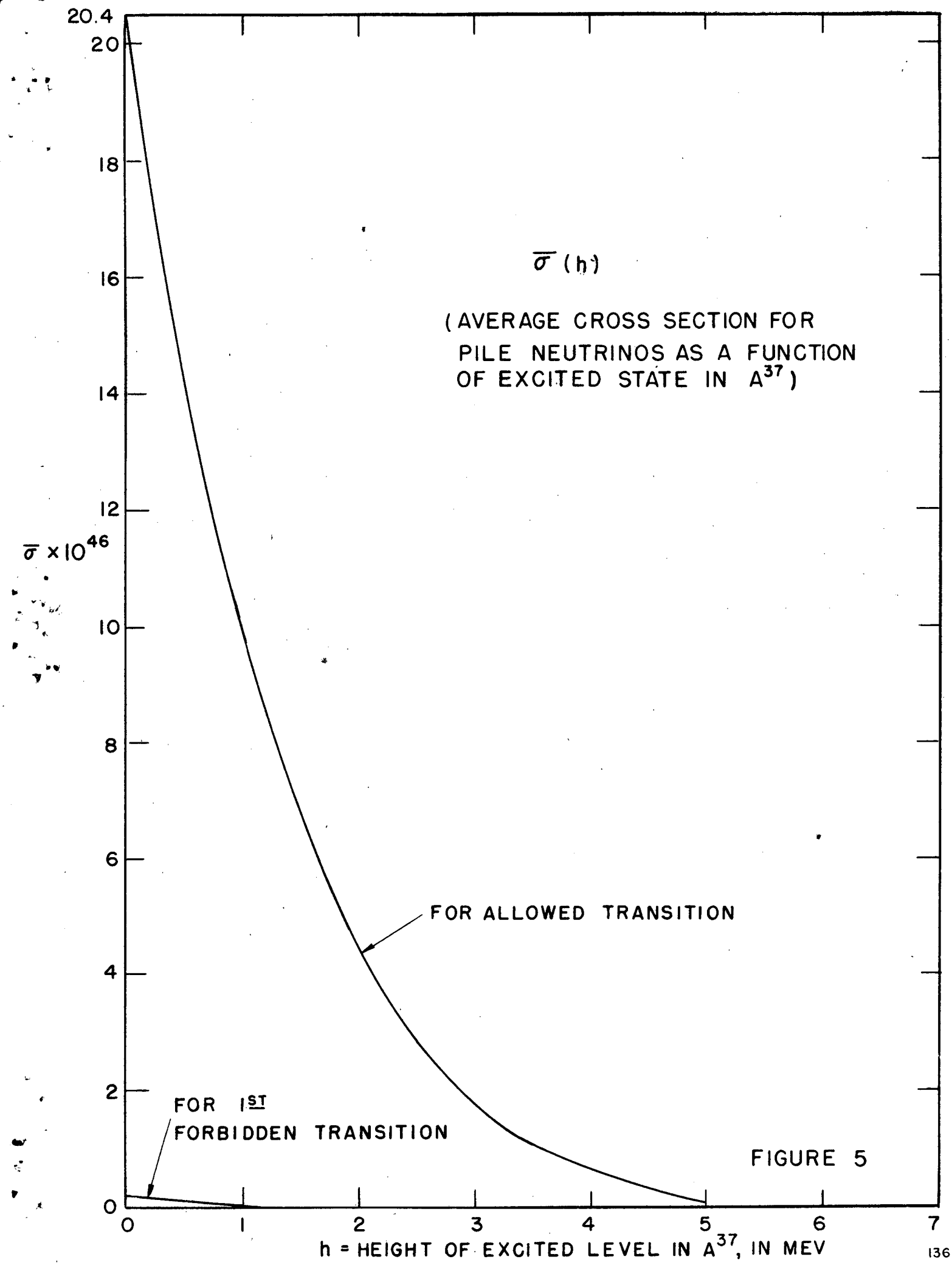
$\overline{\sigma}$ (h)
(AVERAGE CROSS SECTION FOR PILE NEUTRINOS AS A FUNCTION OF EXCITED STATE IN A^{37})
$\overline{\sigma} \times 10^{46}$
20.4
20
18
16
14
12
10
8
6
4
2
0
FOR ALLOWED TRANSITION
FOR 1ST FORBIDDEN TRANSITION
0
1
2
3
4
5
6
7
h = HEIGHT OF EXCITED LEVEL IN A^{37}, IN MEV
13614-1

FIGURE 5