THE TIME SPECTRA OF POSITRONS ANNIHILATING IN SOME ORGANIC LIQUIDS

by

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Abstract

The intensity I_2, and the mean life τ_2 of the long lived component in positron decay have been measured in various organic halides using a fast time to pulse height converter. It was seen in most cases that the intensity of the long lived component was mainly dependent upon the halogen rather than upon the parent molecule. In general, compounds with a chlorine atom showed an I_2 of 14% - 16%, those with a bromine atom showed an I_2 of about 10% and those with an iodine atom showed an I_2 of about 4%.

The value of τ_2 for the chlorides studied varied from 1.10 nanoseconds to 2.04 nanoseconds. An attempt is made to correlate the τ_2 values with certain properties of the molecules.

Chapter I

INTRODUCTION

Since Deutsch (D-51) investigated the annihilation of positrons in gases, many groups have studied the process in liquids. The earliest results were obtained by DeBenedetti and Richings (De-52), and by Bell and Graham (B-53) in 1952 and 1953. Bell and Graham saw that liquids and some solids exhibited a complex decay scheme in which some of the positrons decayed with a mean life τ_1 of about 10^{-10} seconds, and the remainder with a mean life τ_2 of about 2×10^{-9} seconds. Deutsch (D-51) had earlier verified the existence of positronium (a positron-electron pair in a bound state) in gases; Bell and Graham explained their results by postulating the formation of positronium in these materials in spite of the fact that the orbital radius of a positronium atom is usually larger than the intermolecular spacings in condensed media.

According to this theory, positronium can be formed in the triplet state, (parallel spins) or in the singlet state (anti-parallel spins) for which the calculated lifetimes are of the order of 10^{-7} and 10^{-10} seconds respectively. From quantum mechanical considerations it has been shown that the decay from the triplet state must go exclusively via 3 quantum annihilation and decay from the singlet state must be by 2 quantum annihilation. Since Bell and Graham showed that their long lived component decayed by 2 quantum annihilation, they suggested that positronium which originally formed in the

triplet state was being converted to the singlet state by collisions and was then decaying with the short lifetime of 10^{-10} seconds resulting in an effective lifetime of about 2×10^{-9} seconds. In their investigations they found originally that most materials with a τ_2 component that were studied exhibited an I_2 (the percentage of positrons decaying with the longer lifetime) of about 30%. This was due mainly to a chance selection of materials and large errors. By 1957 the results of various workers (G-57) showed that I_2 varied from ~2% to 53%.

Green and Bell (G-57) performed experiments to measure variations in the intensity of the long lived component in water upon adding different concentrations of NO_3^- and NO_2^- ions. They saw that increasing concentrations depressed I_2 from its normal value in water of 21 $\pm$ 5% to 2% without any appreciable change in τ_2 and explained this by postulating the formation of positron compounds such as $e^+ NO_3^-$ in the water. Positrons thus captured would decay with a lifetime similar to that of singlet positronium, i.e. 10^{-10} seconds, and hence would be lost from the τ_2 component of the curve. The addition of other ions however, such as Cl^-, left the I_2 relatively unchanged and the above chemical hypothesis did not hold true.

In 1958, Hatcher, Millet and Brown (H-58) measured the mean life and the intensity of the long lived component in several organic compounds, both liquids and solids. They attempted to find a qualitative correspondence between the different values of I_2 and certain properties of the molecules and found a reasonably consistent correlation between I_2 and the force constant for the weakest bond in the molecules. They could also explain some of their results by postulating that a concentration of negative charge in the molecule would reduce I_2 by causing more positrons to annihilate with bound electrons.

Most of the above mentioned work was done with equipment having a time resolution of about 2 nanoseconds, but recently types of apparatus have been developed having resolutions as small as a fraction of a nanosecond. This has enabled workers to see τ_2 components where it was thought that none existed, and it would appear now that all materials exhibit a τ_2 component.

Chapter II

APPARATUS

Sources:

A Na^{22} source, which emits a positron with a maximum energy of .541 Mev effectively in coincidence with a 1.28 Mev gamma ray, was used in the positron annihilation studies. The Na^{22} was in the form of an queous solution of $Na^{22}Cl$ with a specific activity of 57 μc/cc, obtained from Abott Laboratories in Chicago, Illinois.

Three different preparations of $Na^{22}Cl$ were necessary in order to have positrons decay in water, aluminium and organic liquids.

For the decay of positrons in water, approximately 5 μc of Na^{22} were dissolved in a test tube of distilled water. No difficulty is encountered due to the fraction of positrons escaping from the water and annihilating in the pyrex glass since the decay schemes for positron annihilation in water and pyrex glass are very similar. (G-57).

To obtain some of the prompt coincidence curves, an aluminum sandwich source from A.E.C.L. was used. Other prompt coincidence curves were obtained with a Co^{60} source of about 5 μc.

With the exception of water, $Na^{22}Cl$ was not soluble in any of the liquids studied, so an open source could be used. This consisted of a thin (3 mg/cm^2)

roughened sheet of mica onto which 5 μc of Na^{22} were evaporated over an area of about 1 cm^2. The mica sheet was then lowered into the solution to be studied.

With the apparatus used here, there would be no detectable long lived component for positrons decaying in mica. These would therefore add counts only to the prompt portion of the decay curve for the liquid being studied. Green and Bell (G-57) estimated that less than 10% of the positrons annihilate in the mica in a sandwich source of the thickness used in this experiment, so it is reasonable to assume that less than 5% annihilate in the mica in an open source.

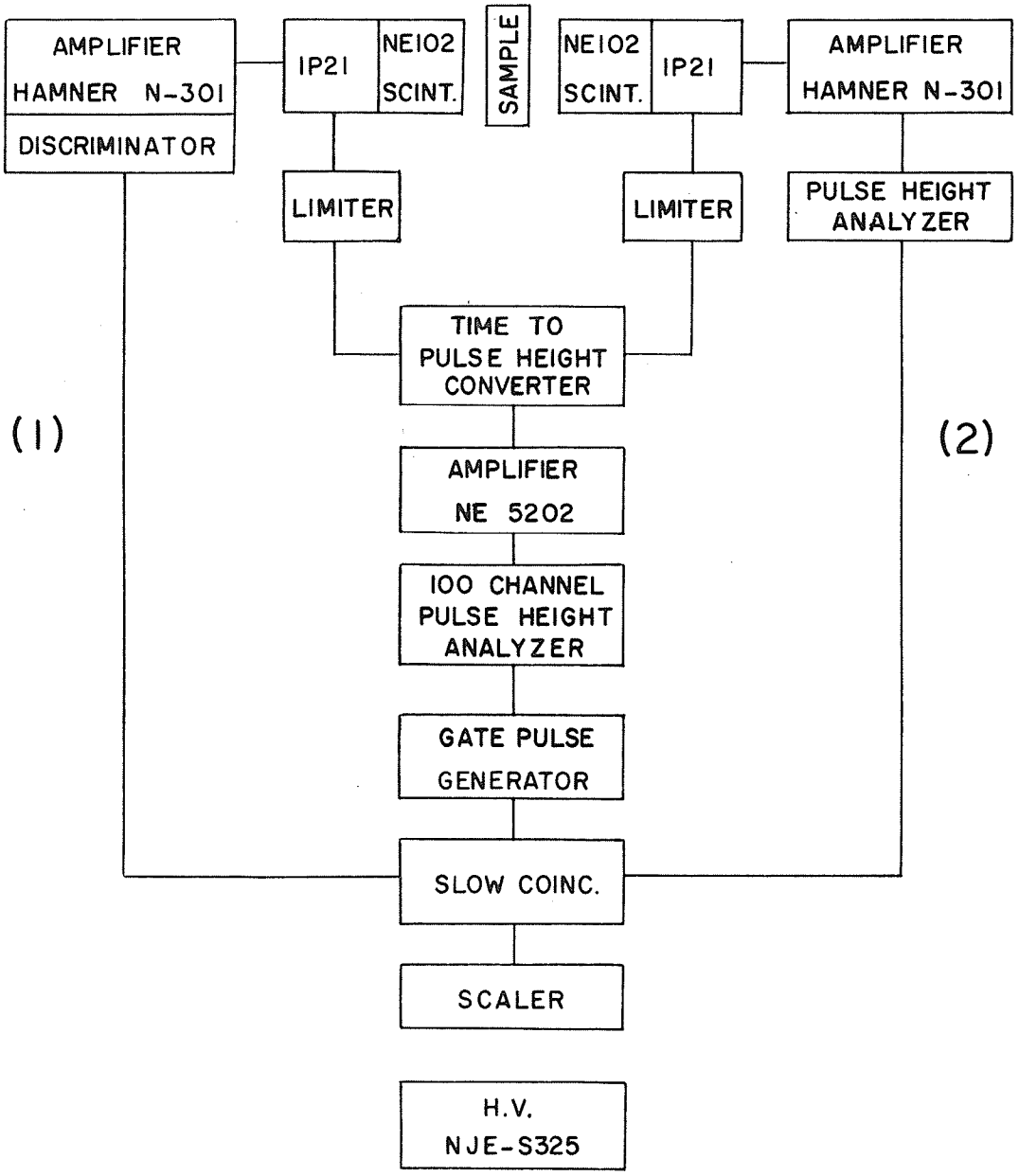
General description:

The function of the apparatus is to measure the time delay between the emission of a 1.28 Mev gamma ray from the source and a .511 Mev gamma ray from the sample. This is a measure of the lifetime of the positron in the sample since a positron is emitted effectively in coincidence with the 1.28 Mev gamma ray, and the appearance of a .511 Mev gamma ray (or rather a pair of such gamma rays correlated at 180°) signifies the annihilation of the positron with an electron. Some 3 quantum annihilation does occur, but it is less than 1/2% of the total and can be ignored.

Fig. 1 shows a block diagram of the apparatus. In order that a count may be registered in the 100 channel

FIG. 1

BLOCK DIAGRAM OF APPARATUS



pulse height analyser, there must be a coincidence in the unit marked 'slow coincidence', and a pulse of sufficient amplitude to enter the kicksorter (after amplification) must come from the time to pulse height converter. The discriminator in the side channel marked '1' is set to pass pulses to the slow coincidence unit proportional to energies greater than .52 Mev, and the pulse height analyser in side channel 2 is set to pass pulses proportional to energies between .2 and .5 Mev; i.e. side channel 1 accepts the compton distribution from 1.28 Mev gammas and side channel 2 accepts the Compton distribution from the .511 Mev annihilation radiation. If the two events occur within 0.5 microseconds of one another, then a gating pulse is sent to the kicksorter from the gate pulse generator. The number of such 'slow coincidences' is recorded on the scaler. Pulses from the 1P21 photomultipliers are sent to their respective limiters where they are changed to pulses of a constant height and length. These shaped pulses go to the time to amplitude converter, where, if they overlap, the amount of overlap is converted to a pulse height. This voltage pulse is then amplified and sent to the kicksorter where it is recorded if there has also been a slow coincidence. The bias is set on the kicksorter so that if the two pulses do not overlap, no count is registered even though a slow coincidence may have occured.

Detectors:

The detectors consisted of plastic phosphors (NE 102) cemented with R 313 bonding agent to R.C.A. 1P21 photomultiplier tubes. The reason for employing a glue rather than silicon fluid will be discussed in Chapter 3. The plastic phosphors were cylindrical in shape, 2 cm. high by 2 cm. in diameter. A portion of the surface was ground to fit the face of the 1P21 photomultiplier. Although the counting efficiency and energy resolution is poor in these plastics, pulses of very short mean time duration ($\sim 2 \times 10^{-9}$ seconds) are produced. Plastic phosphors were chosen instead of stilbene crystals since they can be more easily machined. The plastics were wrapped with aluminum foil to provide diffuse reflection. The detectors were then made light tight by wrapping them with black scotch electrical tape.

The 1P21 photomultiplier tube is superior to conventional end window tubes since it has an internal photocathode and hence short transit time spreads. They also have the advantage of high gain. However, it would probably have been preferable to use a tube such as the R.C.A. 7264 which has a curved photocathode which results in almost spherical geometry. Good optical coupling to the

scintillator is more easily obtained in these tubes than in those with internal photocathodes.

1P21 photomultipliers are normally operated at 900 volts but to ensure as small transit time variations as possible they were operated at 2300 volts. This was the maximum permissible voltage - anything higher resulted in the tubes going into continuous discharge. At this high voltage, noise pulses became excessive, so two of eight tubes available were chosen for best signal to noise ratio which was approximately 4 to 1 for 1.28 Mev gamma rays.

As shown in Fig. 2, a bleeder chain of 220 K resistors placed across a regulated high voltage supply determined the dynode voltages.

Limiters: (J-60)

The performance of the limiters is very critical in this type of fast coincidence circuit for reasons which shall be discussed in the next section. The limiter tube must be a sharp cut-off tube capable of producing a flat-topped anode pulse with a rise time of the order of 1 nanosecond. It must also be able to conduct a large d.c. plate current (15-20 milliamperes) in order that the fast rising pulse have an amplitude of at least 1 volt. A recently

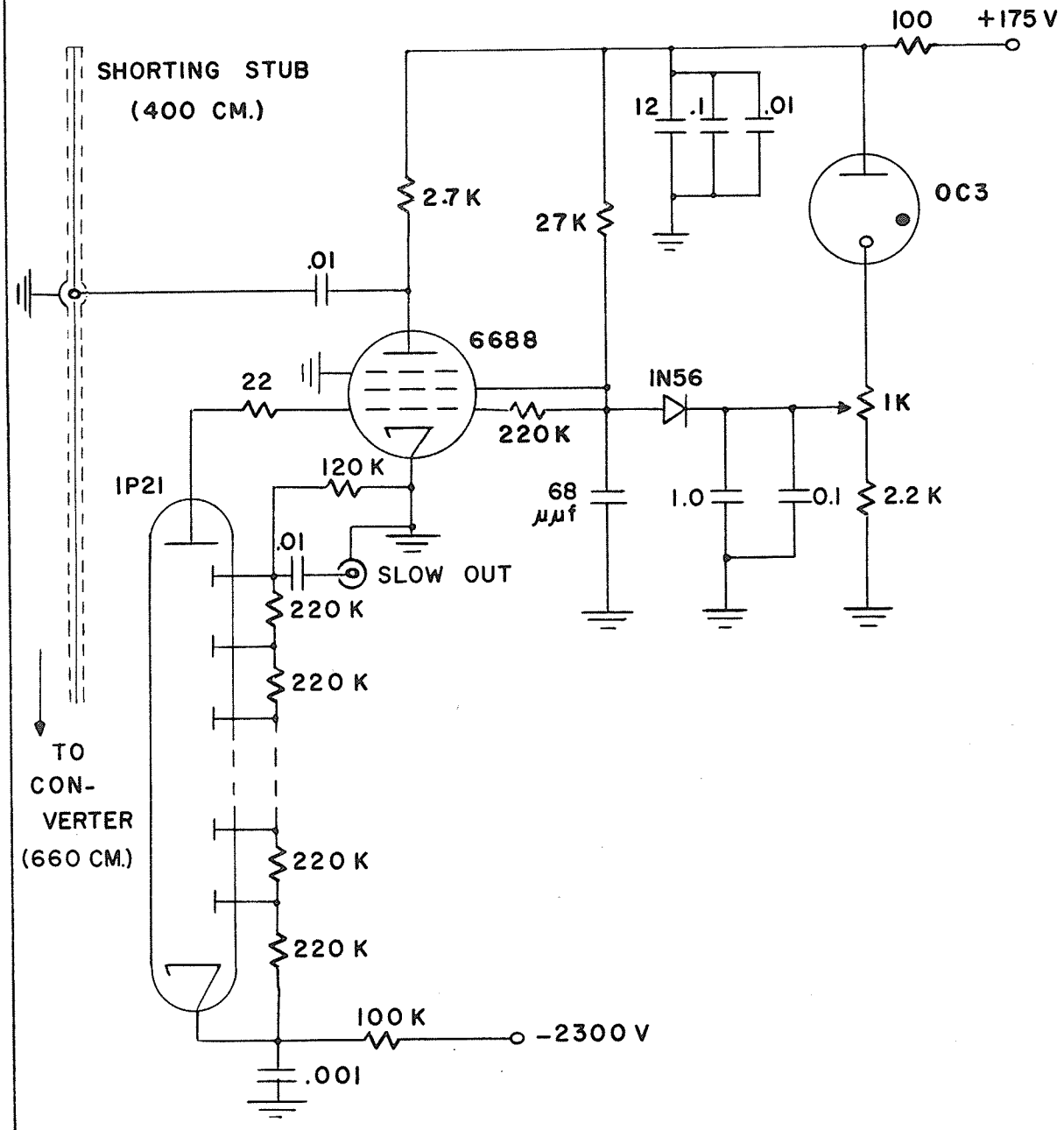
developed tube, the E 180 F,(a Philips tube marketed in North America under the number 6688) is suitable for this application since it has high gain, high current capacity, and a sharp cut-off. The limiter circuit as a whole must be such that the pulses produced are of a constant amplitude and independent of the counting rate.

Fig. 2 shows the circuit diagram for the limiters. The plate voltage is 175 volts which results in a plate current of 15 milliamperes. The screen is run at 70 volts and the screen current is about 4 milliamperes. Under these operating conditions the power dissipation of both the anode and the screen are kept within their rated maxima.

In d.c. operation the circuit has negative feed-back since the grid supply resistor is connected to the screen. If the screen current decreases, indicating reduced tube conduction and decreased plate current, then the screen voltage must rise. This results in an increase in grid current thus increasing the total tube conduction; i.e. connecting the grid supply resistor to the screen tends to stabilize the plate current helping to keep the amplitude of the output pulses constant.

FIG. 2

DETECTOR AND LIMITER



Pulses from the photomultipliers are negative and of the order of 10 volts in amplitude. When such a pulse arrives at the grid of the E 180 F, the tube ceases to conduct and the voltage at the plate increases producing the required fast rising, flat-topped pulse about 2 micro-seconds long. The screen voltage also increases, charging the 68 mmf. decoupling condenser. However, the 1N56 diode is biased 2 volts positively (i.e. the screen voltage supply is adjusted by means of the 1 K potentiometer so as to be 2 volts more positive than the screen itself) so the positive excursion of the screen is limited. Therefore, when the screen reaches the screen supply voltage, the diode starts to conduct and clamps the screen to this voltage. When the tube again begins to conduct, the screen voltage drops to its normal value as the decoupling condenser discharges through the 27 K resistor with a time constant of $RC = 68 \times 10^{-12} \times 27 \times 10^{3} = 1.8 \times 10^{-6}$ seconds. By having a small decoupling condenser, the screen voltage is able to recover in a short time. This aids in reducing counting rate effects.

Another factor minimizing these effects is the use of a negative high voltage supply on the photo-multipliers. This eliminates the necessity of using a

coupling condenser between the anode of the photomultiplier and the grid of the limiter tube.

The value of the grid supply resistor (220 K) is chosen so that the grid circuit is recharged to its normal d.c. voltage (0.5 volts) within approximately 1 microsecond after the end of the photomultiplier pulse. Since the grid is clamped, a fairly constant value of recharge current is supplied to the capacity between the grid circuit and ground after the grid has been charged negatively by the pulse from the photomultiplier. This results in very rapid decay of pulses applied to the grid. For example, a pulse that normally decays exponentially in about 20 microseconds decays at the grid in less than one microsecond. This is therefore another factor which minimizes counting rate effects.

The 2 microsecond long anode pulse travels to the junction of the shorting stub and the cable leading to the time to pulse height converter. Both are RG-7/U 97 ohm co-axial cables. The shorting stub is 400 cm. long; the speed of pulse transmission in this cable is 0.84 c, so a time of 32 nanoseconds is required for the pulse to reach the end of the stub, be reflected negatively and return to the junction. Thus, the resultant pulse to the

converter is one whose length is 32 nanoseconds. Due to the output capacity of the limiter tube (10 mmf.) and stray capacities at the junction, there is a capacitive mismatch resulting in multiple reflections at the junction. By experimenting, it was possible to choose a terminating resistance in the time to pulse height converter that reduced reflections so they did not affect the operation of the converter.

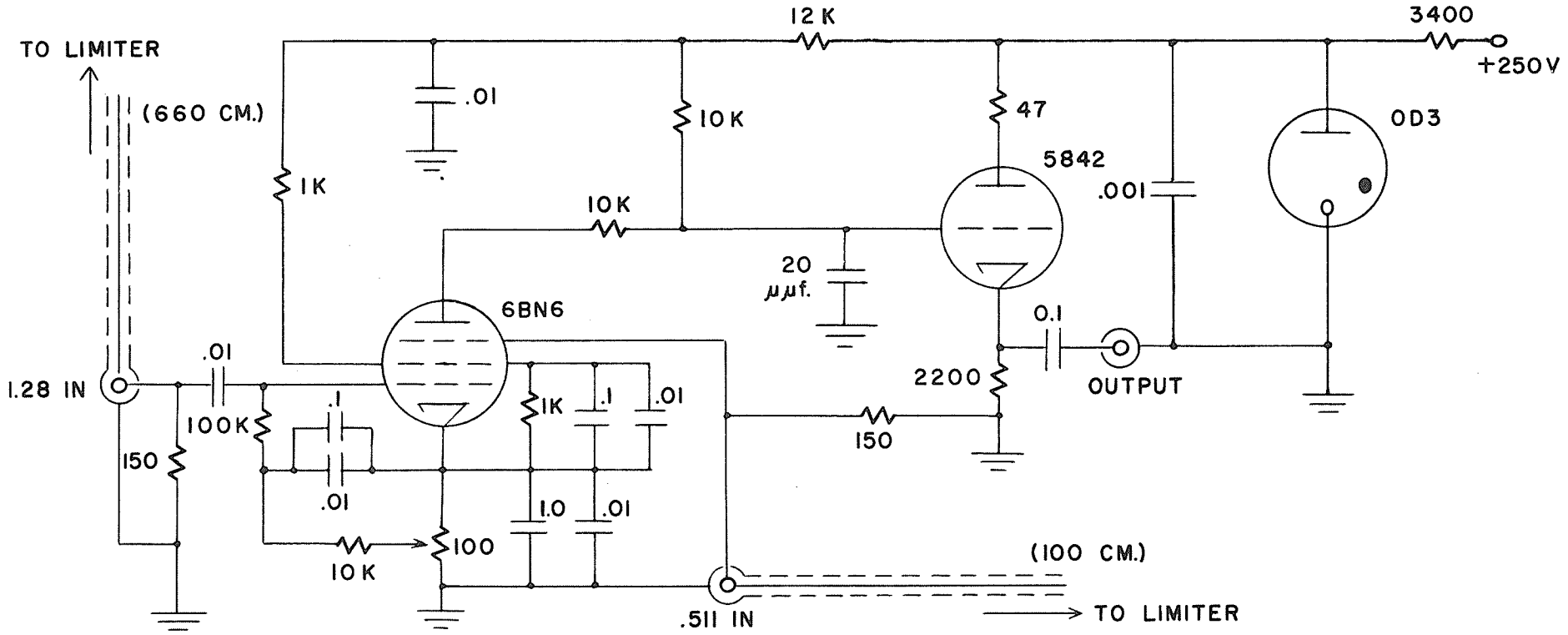
Time to Pulse Height Converter:

The time to pulse height converter is a unit which converts the degree of overlap of two pulses into a voltage pulse; i.e. it converts the time interval between two pulses into a pulse height which can be recorded on a multichannel analyser.

The circuit used here (Fig. 3) is based upon one described by Bell(B-58) but it has several modifications. It employs a 6 BN 6 gated beam tube which has two control grids, g_1 and g_3, called the signal or limiter grid and the quadrature grid respectively. The pulses from the limiter in the 1.28 Mev side channel are applied to the signal grid and those from the limiter in the .511 Mev side channel are applied to the quadrature grid.

FIG. 3

TIME TO PULSE HEIGHT CONVERTER



The electrode voltages on the 6 BN 6 are as follows: anode voltage, E_a = 20 volts; signal grid voltage, E_{g_1} = 0.2 volts; accelerator grid voltage, E_{g_2} = 10 volts; quadrature grid voltage, E_{g_3} = 0 volts and cathode voltage, E_K = 1.0 volts. Therefore, relative to the cathode, the signal grid has a negative bias of 0.8 volts, and the quadrature grid has a negative bias of 1.0 volt. In this application the tube is normally non-conducting. When the grids are driven positive from cut-off, the anode current increases towards a limiting value which is almost independent of further positive excursions of the grids. The grid voltage swing required for the plate current to reach its limiting value depends largely on the electrode voltages. With the low electrode voltages used, a 1.5 volt pulse on both grids is sufficient. However, the maximum pulse amplitude obtainable from the limiters is 1 volt which results in the 6 BN 6 operating in the knee of the E_{g_1} vs. plate current curves when the tube is conducting. In this situation the value of the plate current is still affected by fluctuations in the amplitude of the limited pulses. One method of obtaining a sufficiently large limited pulse would be to use 185 ohm co-axial cable in place of the 97 ohm cable. This would give about a 2

volt pulse but the rise time would be adversely affected, and reflections due to the capacitive mismatch at the junction on the limiter boxes would be increased. If the bias on the control grids was made less negative, then a 1 volt pulse would be large enough to drive the plate current to its limiting value, but then the increase in the plate current (and the decrease in the plate voltage) under pulsed conditions would be too small to be detected and integrated.

The operation of the complete converter circuit is as follows. An appreciable anode current will flow only if both grids are driven positive. Therefore, the tube conducts only when the limited pulses to the control grids overlap, the time during which conduction occurs being determined by the amount of overlap. While the tube is conducting, the voltage at the grid of the 5842 triode decreases with a time constant given by $RC \cong 10^4 \times 20 \times 10^{-12} = 0.2 \times 10^{-6}$ seconds. Since the overlap times measured are about 20 nanoseconds at most, the portion of the exponential decay curve being used is linear to a first approximation. Thus, the amplitude of the pulse at the grid and at the cathode of the 5842 is directly proportional to the length of the anode pulse at the 6 BN 6 which in turn is proportional to the amount of overlap of the limited pulses.