# ANGULAR CORRELATION OF GAMMA RAYS FOR POSITRONS ANNIHILATING IN SOME OXYGEN-LIQUID MIXTURES AND IN FERROMAGNETIC SUBSTANCES

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## ABSTRACT

The angular correlation of gamma rays for positrons annihilating in a number of oxygen-liquid mixtures and in two magnetized substances have been performed. The angular correlation data of $O_2$-liquid mixtures were converted to momentum distributions of the annihilating electron-positron pairs. It was found that the momentum distribution shapes were nearly independent of the amount of dissolved oxygen, while the long life time $\tau_2$ is severely quenched by dissolved oxygen. The quenching mechanism of $^3S$--$^1S$ conversion was rejected and the formation of positronium compounds postulated to account for the experimental angular correlation data.

Preliminary investigations on polarized positron annihilation in magnetized Alnico and $(La_{.70}Pb_{.30})MnO_3$ have been reported. It was confirmed that the shape of the polarization curve for Alnico was in agreement with the shape of polarization in Fe which indicated an antiparallel conduction band polarization.

# CHAPTER 1

## INTRODUCTION

When a positron enters a condensed medium it may annihilate directly with an electron or it may capture an electron to form positronium in the triplet or the singlet state. Triplet or singlet positronium is formed depending on whether the spins of the positron and the electron are parallel or antiparallel. The singlet state annihilates with the emission of two quanta, while the triplet state annihilation is accompanied by a three-quantum emission. Direct annihilations and annihilations from the singlet state of positronium have a mean life time $\sim 1.25 \times 10^{-10}$ seconds. This is known as the $\tau_1$-component. The mean life time of the triplet positronium with a three-quantum annihilation is $\sim 1.4 \times 10^{-7}$ seconds.

Associated with two-quantum annihilation, there is also observed in a condensed medium a second longer life time of the order of $10^{-9}$ seconds. It is known as the $\tau_2$-component. The $\tau_2$-life time is interpreted as arising from "pick off" annihilation from the $^3S$ state ( Dresden 1954; Wallace 1955 ). The so-called "pick off" process may occur in which the positron annihilates by two-quantum

emission with an atomic electron whose spin state relative to it is singlet.

When a positron-electron pair at rest annihilates with the formation of two-photon emission, an energy of $2mc^2$ is released where m is the mass of electron and c is the velocity of light. To conserve momentum, these two photons (each will have a momentum mc) are emitted at $180^\circ$ to each other in the center of mass system. If the annihilating pair has some momentum at the time of annihilation, then the photon pair will be emitted at an angle differing from $180^\circ$ by an amount of the order of v/c where v is the velocity of the center of mass of the annihilating pair. For the low velocities the departure of the angle between the direction of the photons from $180^\circ$ is proportional to the component of momemtum of the annihilating pair which is parallel to the bisector of the propagation directions. Thus, one may measure the angular distribution of annihilation photons and convert this to a momentum distribution of the annihilating positron-electron pairs ( Stewart 1957 ).

The study of angular correlation of two-photon annihilation of positrons in matter has been investigated

by many investigators and has become a useful tool in study of atomic and molecular properties. The original measurements of the angular correlation of photons from positron annihilation were made by Beringer and Montgomery in 1942. The first detailed theoretical and experimental investigations employing far greater precision have been done by de Benedetti et al. in 1950. Following this, both Warren et al. ( Argyle and Warren 1951; Warren and Griffiths 1951; and Erdman 1955 ) and Maier-Leibnitz (1951) also measured the angular correlation of photons from annihilations in various elements and compounds.

Most of the extensive measurements with metals and alkali halides have been done by Stewart et al. ( Stewart and Green 1955; Green and Stewart 1955; Stewart 1955;1957; Stewart and Pope 1960; Donaghy and Stewart 1964 ) and by de Benedetti and co-workers ( Lang, de Benedetti, and Smoluchowski 1955; Lang, de Benedetti 1957 ). It was found that the half-width of the angular distribution was a good measure of the Fermi energy of light metals, and that the angular distributions can be converted to the momentum distributions of the annihilating positron-electron pairs. Page and co-workers ( Page, Heinberg, Wallace,

and Trout 1955; Page and Heinberg 1956 ) also measured positron annihilations in organic and amorphous substances. They have shown that angular correlation measurements give information about the mechanism of positron annihilation in solids and about the electrons of the solid with which the positrons annihilate.

In the study of the factors affecting the formation and destruction of positronium in matter, de Zafra ( 1958 ) has reported some measurements of the angular correlation of photons from positron annihilation in condensed materials under varying conditions of temperature, pressure and phase, and in the presence of certain types of impurities. It was found that the underlying cause of the density-temperature effect seemed to be simply a variation in fraction of positrons forming positronium. The influence of paramagnetic ions in causing $^3S \longrightarrow {}^1S$ conversion in positronium by electron exchange was also investigated.

In 1960, Trumpy has also measured the angular correlation of photons from positron annihilation in aqueous solutions. It was confirmed that the amount of singlet positronium formed is influenced by two processes: a reduction of positronium due to electron capture by oxidizing substances

and an increase of the $^3S \longrightarrow {}^1S$ conversion due to electron exchange with paramagnetic ions. The oxidation potential of positronium was found to be very nearly zero, and the conversion rate was proportional to the number of unpaired electrons on the dissolved ions.

Kerr (1964) has measured the angular correlation of photons from positron annihilation in a number of organic liquids with increased experimental accuracy. The angular correlation data have been converted successfully to momentum distributions of the annihilating pairs in contrast to previous experiments in liquids where the experimental accuracy had not been sufficient to allow such a conversion. It was found that there are two momentum components which appear in the momentum distributions whenever positronium is formed. It was confirmed that the intensity of the low momentum component is equal to one third of the intensity of the long lived component. He also observed that the high momentum component was a direct measure of the momentum distribution of the electrons involved in the annihilation process.

Recently some measurements of the time spectra have been reported ( Lee and Celitans 1965; Kerr, Cooper, and

Hogg 1965 ) that the long lifetime ($\tau_2$) for positron annihilation in some organic liquids is severely quenched when oxygen is dissolved in the liquid. This has been interpreted previously as being due to $^3S \longrightarrow {}^1S$ conversion by paramagnetic oxygen. Hogg et al. ( Kerr, Cooper, and Hogg 1965) has shown that the conversion mechanism was not responsible for the quenching of $\tau_2$ in hexane.

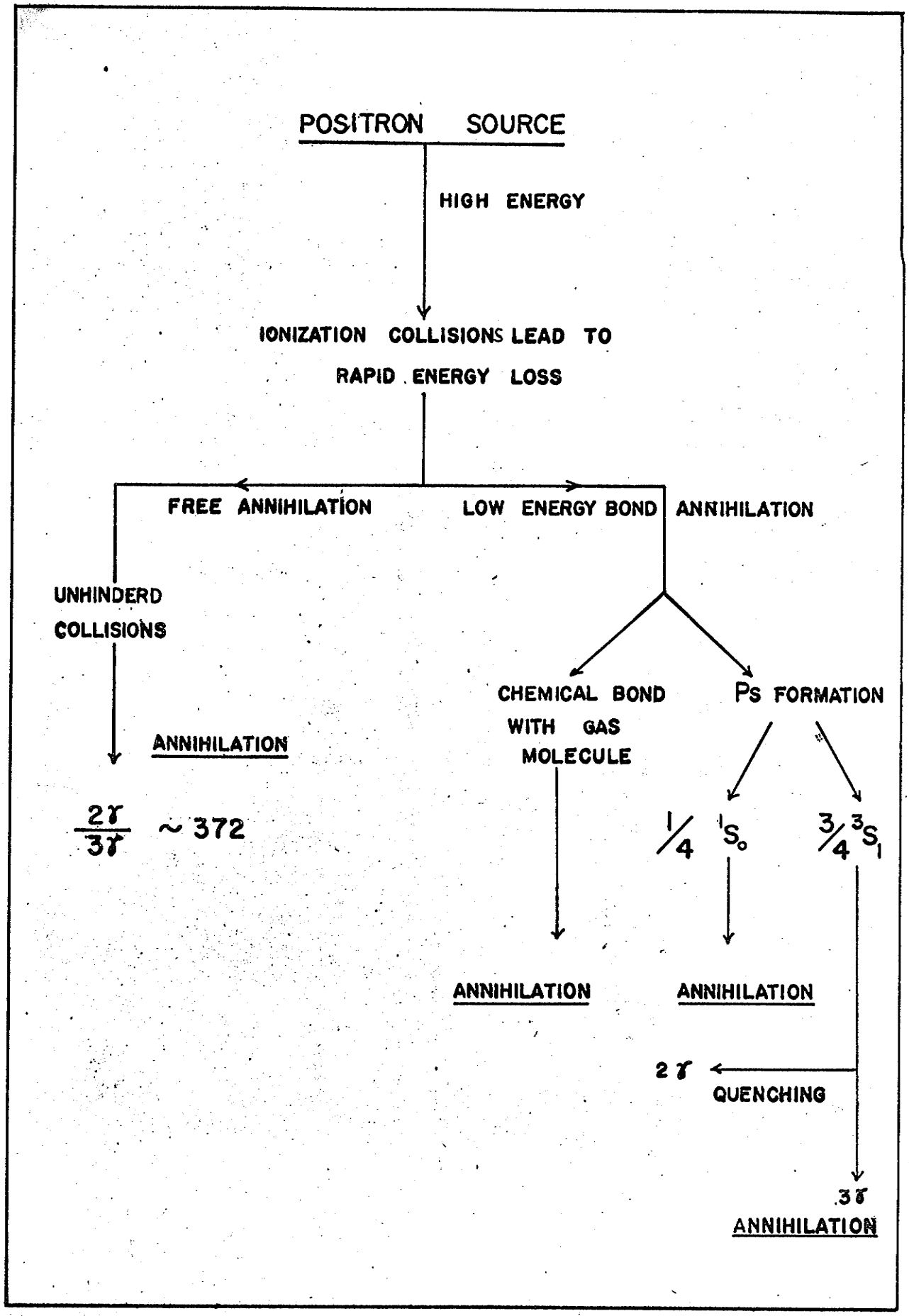
In this work it will be shown again that the quenching of $\tau_2$ in a number of $O_2$-liquid mixtures is not due to the $^3S \longrightarrow {}^1S$ conversion. It will be interpreted as being probably due to the formation of positronium compounds.

In the last part of this work, some preliminary investigations on polarized positron annihilation in ferromagnets will be reported. The positron annihilation angular correlation method was first used by Hanna and Preston (1958) to investigate the momentum distribution of electrons responsible for ferromagnetism. In order to obtain information concerning the state of polarization of these electrons, recently some investigators ( Mijnarends and Hanbro 1964; Berko and Zuckerman 1964 ) have repeated these measurements in iron and nickel. It was found that the conduction electrons in ferromagnets were polarized

antiparallel to 3d electrons, and that the interpretation of the polarization experiments was somewhat model dependent. A series of experiments on a perovskite ferromagnetic substance ($La_xPb_{1-x}MnO_3$) without conduction electrons have been performed. The same ferromagnetic substance with various conductivities (by changing temperature) is presently under investigation in our laboratory.

FIG 1

FLOW DIAGRAM FOR POSITRON ANNIHILATION


POSITRON SOURCE
HIGH ENERGY
IONIZATION COLLISIONS LEAD TO
RAPID ENERGY LOSS
FREE ANNIHILATION
LOW ENERGY BOND ANNIHILATION
UNHINDERD
COLLISIONS
ANNIHILATION
2γ / 3γ ~ 372
CHEMICAL BOND
WITH GAS
MOLECULE
Ps FORMATION
1/4 1S0
3/4 3S1
ANNIHILATION
ANNIHILATION
2γ
QUENCHING
3γ
ANNIHILATION

# CHAPTER 2

## APPARATUS

The angular correlation apparatus consisted of two main parts---the mechanical and electronics components. A detail of arrangements of the apparatus will be described as follows.

### Mechanical:

A diagram of the mechanical parts of the angular correlation apparatus is shown in Figure 2. The basic mechanical features are the source and sample housing, with surrounding collimating slits and two gamma ray detectors with their collimating slits. All these components were mounted on two parallel 3" by 6" aluminium I-beams approximately 20 feet long. The two gamma tay detectors were mounted on two ends of the I-beams. One detector was fixed and the other was movable. The source and the sample were shielded in a lead castle, being placed on the middle of the I-beams, with collimating slits facing each detector.

The widths of the collimating slits of the lead castle were 0.1" and 0.4", facing the fixed detector

and the movable detector respectively. The reason for a wider slit, on the side of the movable detector, is that the movable detector had to be permitted to see the sample directly from a variety of positions, The purpose of these collimating slits was to reduce the amount of scattered radiation reaching the detectors as well as to shield the detectors from the source.

The fixed gamma ray detector ("B" in Fig. 2) and its collimating slits were mounted on a brass plate 265cm from the source. This detector was rather heavily shielded by 3" thick lead blocks to cut down the accidental background rate from scattered gamma rays and from other sources in the laboratory.

The movable detector ("A" in Fig. 2) and its collimating slits were mounted on a steel plate, placed 265cm on the other side of the sample, driven by a worm screw between a set of linear rails. Here a "Slo-Syn" 600 ounce-inch reversible motor was used.

The collimating slits of these detectors were made from 2½" thick lead blocks. This thickness was sufficient to stop most of all gamma rays from this

experiment. The width of slits could be changed by inserting metal shims of the appropriate thickness between the lead blocks.

To keep the collimating slits of the detectors aligned towards the sample as the movable detector moved to different positions, an aluminium beam, which was reinforced by a set of three steel cables to both ends of the beam, was extended from the front of the rotatable steel plate to a pivot point directly under the sample housing on a vertical axis through the sample. To allow for the slight increase in the distance from the detector to the pivot point as the detector was moved off the center position, the end of the aluminium beam was permitted to slide freely in a hole in the pivot shaft.

To define the position of the sample and align the apparatus a fine wire was stretched between the slits of the two detectors which were in the position of 180 degree line. The position of the sample was adjusted so that its face was just in contact with the wire. Checks were made periodically on the alignment, to guard against any accidental shifting of the equipment. (If the slits were shifted slightly out of line, a change in the peak

position of the angular correlation curves would be observed. But such a change would not affect the final experimental results.)

A check was also made on the constancy of the pitch of the worm screw by measuring the distance that the whole slit system of the movable detector moved with each rotation of the screw, The slit system moved 2.54mm with each rotation. A maximum variation of 0.03mm of one rotation was obtained, but a deviation of this magnitude would not affect the experimental results noticeably.

Electronics:

A block diagram of electronics is shown in Fig. 3. The detectors were Integral Assembly model 16MB4/A-X and consisted of 4" diameter, 1" thick NaI (Tl) crystals mounted on 5018 HB photomultipliers with mu-metal shields. A positive high voltage (1000 volts) was provided for photomultipliers by a Hamner N401 high voltage supply. Negative pulses which have the order of one volt in amplitude, from the cathode followers in the detector heads were fed to simple amplifiers (Kerr 1964) through 1059 Amphenol coaxial cable, and were amplified about eighteen times and shortened to approximately 1 microsecond.

The power for the amplifiers and cathode followers in the detector heads was provided by a highly regulated 250-volt D.C. power supply based on a National Bureau of Standards design.

Pulses from the amplifiers were fed to single channel pulse height analysers set to select gamma rays in the energy range between 0.1 and 0.5 Mev. The fast rising narrow pulses (0.2 micro-seconds) were sent to a coincidence unit which had a resolving time of less than 0.5 microseconds. The pulse height analysers and the coincidence unit have been described previously (Naqvi 1961). A Technical Measurement Coorporation Model SG-3A scaler was used to record the number of coincidences.

When 1000 counts have been accumulated on the scaler, through the "automatic unit" (de Zafra 1958; Kerr 1964), the track motor is started and moves the movable detector to its next position, simultaneously, the scaler stops counting as the motor is running, and the scaler starts counting again as soon as the motor stops while the detector has been driven to its new position. When the detector moves to the predetermined end of its run in either direction (eg.12 mr in each

side), it will reverse the direction automatically by reversing switches in the automatic unit system. A Simplex Time Printer Model ET-100 was used to print out the time interval during each 1000 counts of coincidences.

All the electronic instruments were powered by a model 2000 S Sorensen A.C. Voltage regulator.

Preparation of Positron Source:

A 12 millicuries $Na^{22}$ source in the form of NaCl solution obtained from the Radiochemical center, Amersham, England. The source had a high specific activity, 3mc/mg, and was dissolved originally in 6ml. of water.

The source preparation was accomplished as follows: a ½" diameter plastic button on which the $Na^{22}$ was to be deposited was prepared by machining shallow, concentric circular grooves on it. The source was evaporated from solution, drop by drop, on the plastic button mounted on the end of a ½" diameter, 10" long plastic rod, and then covered with a thin piece of mica ($2mg/cm^2$) sealed around the sides of the button with epoxy resin glue. It was estimated that about 10mc of the source was deposited on the plastic button as the evaporation process was finished.

The remainder was left in the original source container, in syringe and in hypodermic needle. Unfortunately, this source was broken by an accident, unexpectedly, during the time of moving it from the radiochemical laboratory in which the source was prepared to our laboratory. The surface of the source was covered again by another piece of mica. The total thickness of the two pieces of mica which had covered the source was about 4mg/cm$^2$. This thickness of the source covering was still thin enough to obtain a reasonable yield of positrons. But the yield of positrons obtained, was much lower than what we had expected. Soon, it was found that this was due to the self absorption in the source since the salt crystals of $Na^{22}$ were accumulated in one area of the surface of the plastic button during the accident. The source was then opened and redissolved in a small amount of water and deposited on a new plastic button which had been prepared in the same manner as the previous one except the grooves on it were a little more shallow. After the process of transfer was completed, it was estimated that about 20% of activity was lost and appoximately 8mc of $Na^{22}$ was deposited on the new plastic button.

The new plastic button, covering with 2.5 $gm/cm^2$ thin mica, was mounted on a plastic rod, and then inserted into a brass tube in the side of the source castle and was held in place by a locking screw. To achieve the maximum counting rate, the source was placed as near the sample as possible without the source becoming directly visible to both detectors through the crude collimating slits of the source castle.

FIGURE 2

MECHANICAL PART OF THE ANGULAR CORRELATION APPARATUS

A. Movable detector

B. Fixed "

C. Positron source

D. Sample tank

E. Reversing switches

F. Cam operated switch

G. Motor

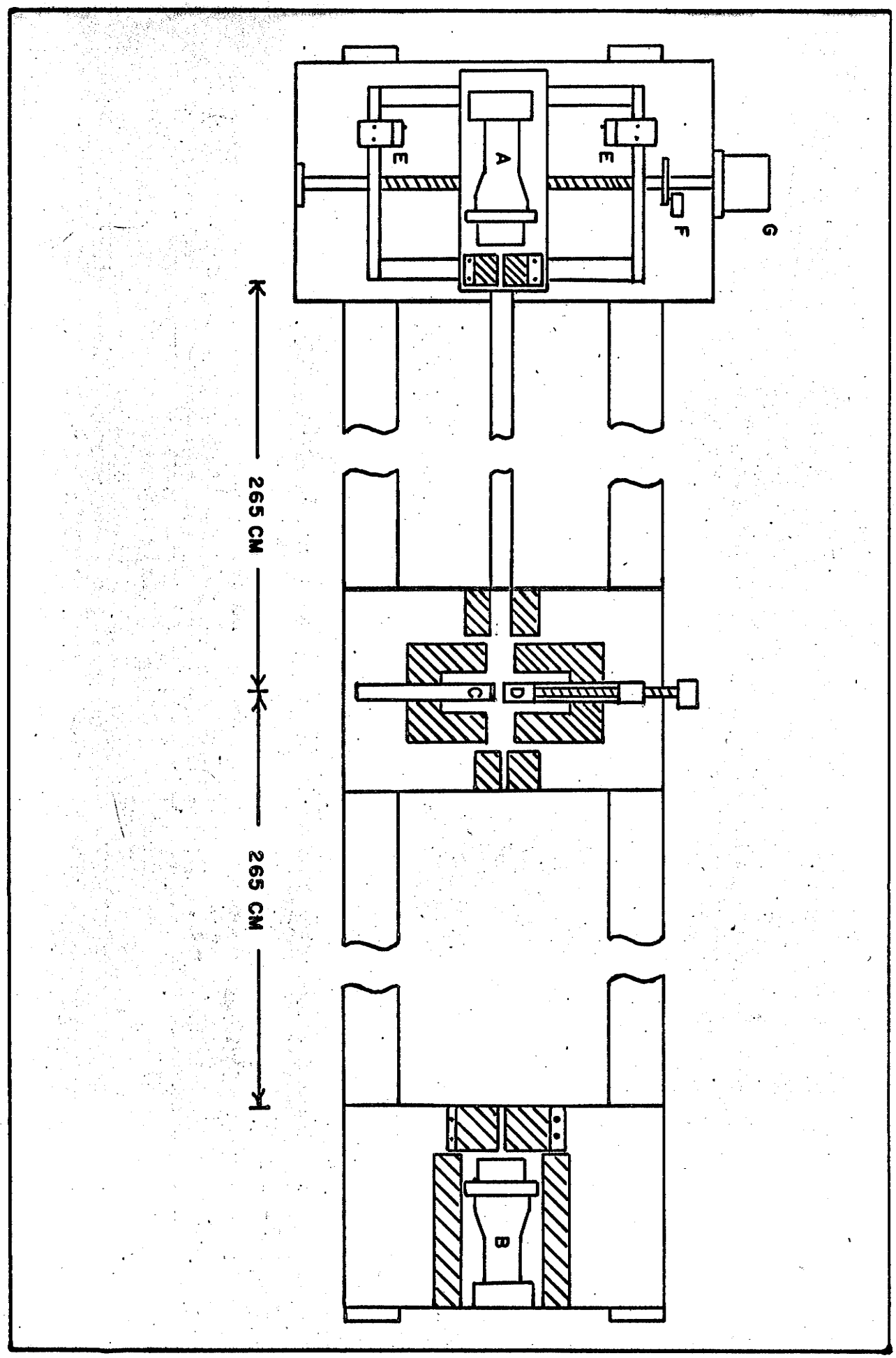
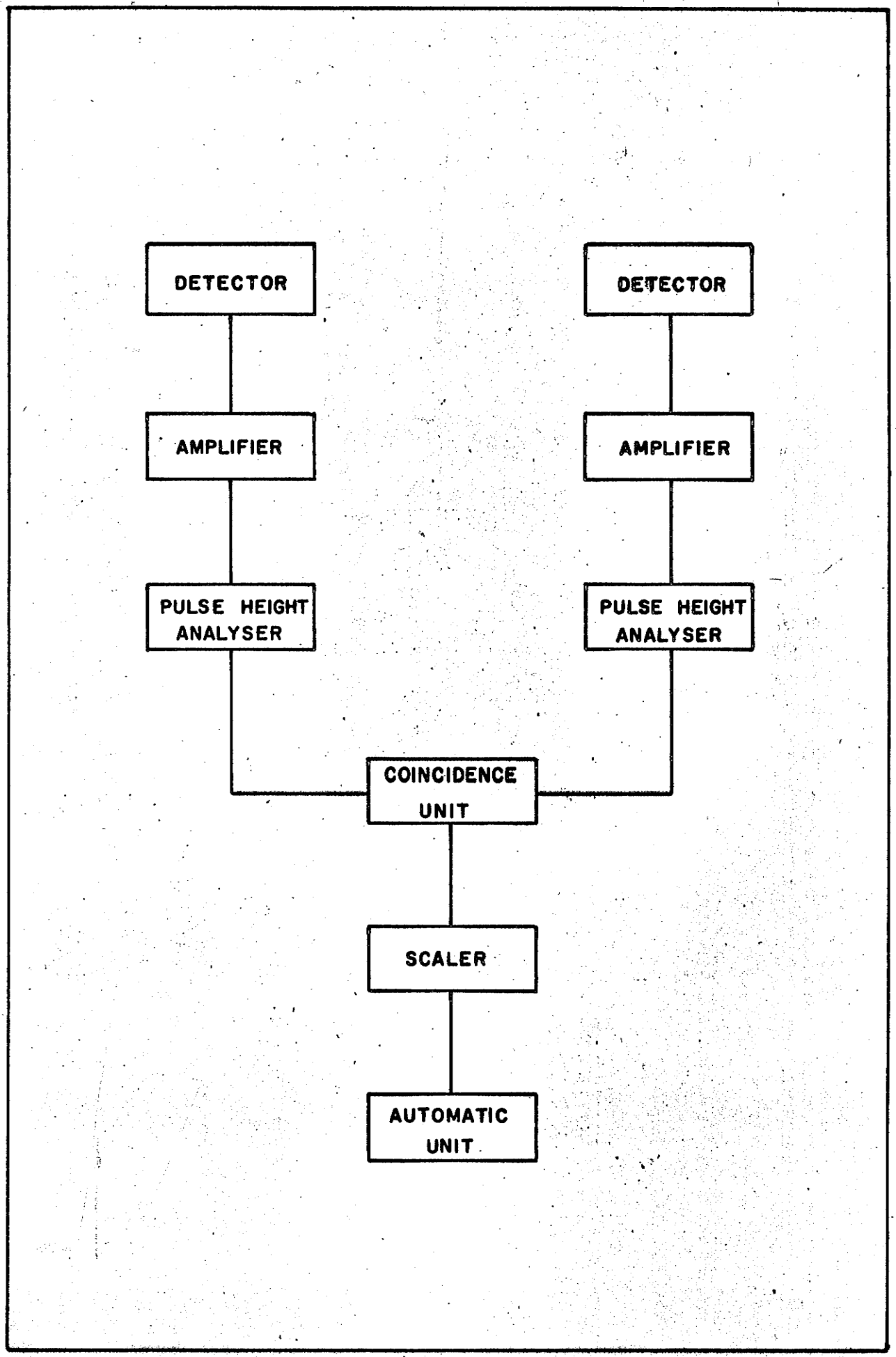

A
B
C
D
E
E
F
G
265 CM
265 CM

FIGURE 3

BLOCK DIAGRAM OF THE ANGULAR CORRELATION APPARATUS


DETECTOR
DETECTOR
AMPLIFIER
AMPLIFIER
PULSE HEIGHT ANALYSER
PULSE HEIGHT ANALYSER
COINCIDENCE UNIT
SCALER
AUTOMATIC UNIT

# CHAPTER 3

## POSITRON DECAY IN $O_2$-LIQUID MIXTURES

### Data Accumulation and Analysis

Two-photon angular correlation measurements were performed for oxygen-saturated, air-saturated, and degassed samples of the following liquids: hexane and two of its halogen derivatives (ie. fluorohexane and chlorohexane), distilled water, and $NaNO_3$ solution in water (1mole/liter). The oxygen-saturated process was accomplished simply by bubbling oxygen through the sample under investigation. The oxygen concentration for this case was estimated to be about 200cc/liter, while for the air-saturated case it was 40cc/liter (Kerr, Cooper, and Hogg 1965). The degassed process was accomplished by the standard vacuum freeze-thaw technique. The degassed sample was analyzed mass spectrometrically and was found to contain less than 1% of the amount of oxygen present in the air-saturated sample.

All these samples were contained in a small brass tank with a 3/4" vertical face covered with a thin mica sheet (1.5mg/cm$^2$) so that more than 95% of the incident

positrons could penetrate through the mica-window and annihilate in the liquid sample. The slits in front of the two detectors subtended an angle of 0.60 milliradians. An angular range about 12 milliradians on either side of the 180° line was set for taking the angular correlation curves. At each angle, approximately 20,000 counts were accumulated.

To determine the background counts in the angular distributions, a brass ring which was made in the manner of having the same diameter and the wall thickness of the sample tank and covered with a same thickness of mice sheet was used to perform several runs in place of the sample. The resulting back-ground distribution was fairly broad with a peak at the center. In addition to this background, the chance coincidences which contributed a flat background of approximately 3 counts per minute was also taken into account.

Stewart (1957) has shown that in the experiment with an apparatus having parallel slit geometry, the angular distribution of the annihilation photons can be converted to the momentum distribution as

$$N(p)=a_1\theta\frac{dC(\theta)}{d\theta} \qquad (1)$$