

POSITRON ANNIHILATION IN MOLECULAR LIQUID-SOLID SYSTEMS

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A Thesis  
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the Faculty of Graduate Studies  
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In Partial Fulfillment  
of the Requirements for the Degree  
Doctor of Philosophy

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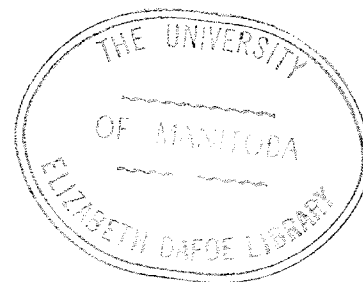
by

Alexander M. Cooper

Winnipeg, Canada

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## ABSTRACT

A fast-slow time sorting system, with day to day resolution of 300 picoseconds, has been constructed to measure positron lifetimes and intensities in Ammonia, Cyclohexane, Methane and Butane. For each substance lifetimes and intensities have been studied over a range of temperatures including in each case a liquid-solid phase change and in the case of Cyclohexane a solid-solid phase change as well.

The data were analyzed using a multi-exponential computer program and the parameters  $\tau_2$ ,  $\tau_1$ ,  $I_2$  and  $I_1$  extracted for each substance at the various temperatures.

The experimental  $\tau_2$ 's obtained for Ammonia and Methane over the entire temperature range studied (including the phase transitions) agree very well with the theoretical values calculated from the Free Volume Model.

The experimental  $I_2$ 's obtained show the mechanism of positronium formation proposed by Brandt and Spirn to be incorrect and an alternate mechanism is postulated which explains the experimental  $I_2$ 's.

A study of the solid-solid phase transition in Cyclohexane yielded a large discontinuity in  $\tau_2$  at the transition temperature and points up the value of the positron probe in exploring phase transitions of this type.



## CHAPTER I

### INTRODUCTION

#### 1.1 The Discovery of Positrons

The positron as a particle was first predicted by Dirac in 1930. His prediction arose from the relativistic wave equation for electron energy:

$$E = \pm \sqrt{m_0^2 c^4 + p^2 c^2}$$

where  $E$  is the total energy of the electron,  $m_0$  its rest mass,  $p$  its momentum and  $c$  the speed of light. Since this equation had both negative and positive energy solutions with the negative solution corresponding to the case of an electron with a charge of  $+e$ , Dirac postulated the existence of positrons or "holes." The particles thus predicted were first detected in 1932 by Anderson in cloud chamber photographs of Cosmic Ray showers. Anderson discovered the particles which he named "positrons" when he applied a magnetic field across his cloud chamber and found particles with electronic mass but positive electronic charge..

With the advent of artificial radioactivity positron sources have become readily obtainable from a number of radioactive isotopes.

## 1.2 Characteristics of Positron Annihilation

The positron is an elementary particle which along with electrons, muons and neutrinos belongs to the class known as Leptons. Since the positron is the antiparticle of the electron the two particles can thus annihilate as a pair to form gamma rays.

The spin of the positron like that of the electron is equal to  $\frac{1}{2}$  and as with other particles of  $\frac{1}{2}$  integer spin positrons obey Fermi-Dirac statistics.

When positrons and electrons collide annihilation can take place with the emission of one, two or three photons. However in the case of annihilation with the emission of only one photon a third body must be present to absorb the recoil momentum and thus conserve the momentum of the system.

To understand two and three photon annihilation we consider a positron-electron pair in an S state. This is sufficient since in higher orbital angular momentum states the positron and electron wave functions do not overlap sufficiently for appreciable annihilation to occur. The positron-electron pair can exist in one of two substates, the singlet state  $^1S_0$  with spin zero and the triplet state  $^3S_1$  with spin one. Now according to Yang (1950) the appearance of two photons upon annihilation is only possible for annihilation from the singlet state. It is known that the relative intrinsic parity for a particle-antiparticle pair is negative and thus for the

positron-electron pair  $P_1 = (-1)$ . Then the charge conjugation for the system is

$$P_C = P_1 P_S = (-1)(-1)^{S+1} = (-1)^S$$

where  $P_S$  is the spin parity of the system. Therefore the charge conjugation is positive for the singlet state ( $S=0$ ) and negative for the triplet state ( $S=1$ ). Now since the charge conjugation of a photon is negative, for a system of  $n$  photons  $P_C = (-1)^n$ . Thus if charge conjugation is invariant in electromagnetic interactions, annihilation from the singlet state ( $^1S_0$ ) must be accompanied by two photons while annihilation from the triplet state ( $^3S_1$ ) must be accompanied by an odd number of quanta (in this case 3 quanta).

This feature of positron-electron annihilation has led to a search for charge non-conservation through the study of three photon decay of the singlet state ( $^1S_0$ ) (A. P. Mills and S. Berko 1967).

A final feature of the annihilation process is the probability of occurrence of each of the possible decay modes. Ore and Powell (1949) have shown that the ratio of the three photon to two photon cross section is  $1/372$  and Goldanskii (1968) has shown that the one photon rate will be more than two orders of magnitude less than the three photon rate.

### 1.3 Discovery and Properties of Positronium

The possible existence of a bound positron-electron

system was first postulated by Mohorovicic in 1934. In 1945 Ruark named this bound system positronium and then in 1951 Deutsch carried out experiments confirming its existence.

The positronium atom is analogous to the hydrogen atom except that its reduced mass is one-half of the electron mass. Thus from simple Bohr theory one finds the Bohr radius for positronium is equal to

$$r_p = \frac{2n^2a_0}{me^2Z^2} = 2a_0 = 1.06 \text{ Angstroms}$$

while in the same way the ground state energy becomes  $E = \frac{e^2}{2r_p} = \frac{e^2}{4a_0} = 6.77 \text{ eV}$ .

There are two ground state configurations for positronium; the triplet state (ortho-positronium) and the singlet state (para-positronium) with the spins being respectively parallel and anti-parallel. Since the triplet state has three magnetic substates ( $m = 1, 0, -1$ ) while the singlet state has only one ( $m=0$ ) the statistical weight of the triplet state is three times that of the singlet state. Thus triplet positronium will be formed three-quarters of the time and singlet positronium will be formed one-quarter of the time. Following the argument of the last section triplet positronium will undergo decay by three quanta annihilation while singlet positronium decays by two quanta annihilation. The lifetimes of triplet and singlet positronium annihilating in vacuum have been calculated as  $1.4 \times 10^{-7}$  seconds and  $1.25 \times 10^{-10}$  seconds (Ore and Powell 1949).

In the formation of positronium, high energy positrons are degraded in energy by various processes such as ionizing and excitation collisions until they reach an energy at which positronium formation becomes favorable. This energy lies within the so-called "Ore Gap" first proposed by A. Ore in 1949. The "Ore Gap" which is controlled by the parameters of the media under investigation and the way in which Ore's theory predicts formation probabilities will be discussed in Chapter V.

#### 1.4 Experimental Methods of Studying Positron Annihilation

There are three experimental methods by which information on positron decay may be gained. The first of these is through the measurement of the triplet annihilation rate by the use of a triple coincident circuit. Since theory predicts that the ratio of three  $\gamma$  to two  $\gamma$  annihilation for free positrons will be 1/372 whenever a rate larger than this is found we have proof of positronium formation. Unfortunately this method cannot distinguish between the case where positronium is not formed and the case where the triplet positronium is quenched by various mechanisms such that it decays by two photon annihilation. Therefore at the present time this method is used only infrequently.

The second method of studying positron annihilation is through the measurement of the mean lifetimes of the annihilating positrons. In this type of experiment a nuclear  $\gamma$  ray is used as a start signal and the subsequent annihilation

radiation is used as a stop signal for a coincidence circuit. It was through experiments of this type that Deutsch in 1951 first showed the existence of positronium in gases. Two types of information are found in experiments of this kind, first the decay rates for the various processes by which positrons can annihilate and second the percentage of positrons decaying by each of the different modes. Since in previous work on condensed media the long lifetime observed has been attributed to triplet positronium this method then allows one to measure directly the amount of positronium formed in a system. Thus by use of the lifetime method one is able to study the effect changes in a given system have on both decay rates and the relative abundance of positrons decaying by the different decay modes.

The third method of measurement is by the technique of angular correlation of the two photon annihilation radiation. When a positron-electron pair at rest annihilates with the emission of two photons an energy of  $2 mc^2$  is released. Since momentum must be conserved these two photons each having a momentum  $mc$  must be emitted at 180 degrees to one another in the centre of mass system. Now 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 degrees by an amount of the order  $v/c$ , where  $v$  is the velocity of the centre of mass of the annihilating pair. Thus by measuring the

departure of the emitted photons from 180 degrees it is then possible to find the momentum distribution of the annihilating pairs. In recent years the analysis of the raw angular correlation data into momentum distributions has been done mainly by electronic computer (S. Y. Chuang 1968).

Figure 1-1 illustrates a typical parallel slit angular correlation apparatus.

### 1.5 Factors Influencing the Annihilation of Positronium

Since the initial discovery of positronium considerable work has been carried out in studying the various factors which effect the lifetime of positronium. Thus positronium lifetimes have been studied as a function of;

Temperature: R. E. Bell and R. L. Graham 1953, H. S. Landes, S. Berko and A. J. Zuchelli 1956, W. Brandt, S. Berko, and W. W. Walker 1960, W. Brandt and I. Spirn 1966

Pressure: R. K. Wilson, P. O. Johnson and R. Stump 1963

Crystallinity: S. J. Tao and J. H. Green 1965

Irradiation: G. Fabri and E. Germagnoli 1962, D. Fabri, G. Poletti, G. Randone 1966

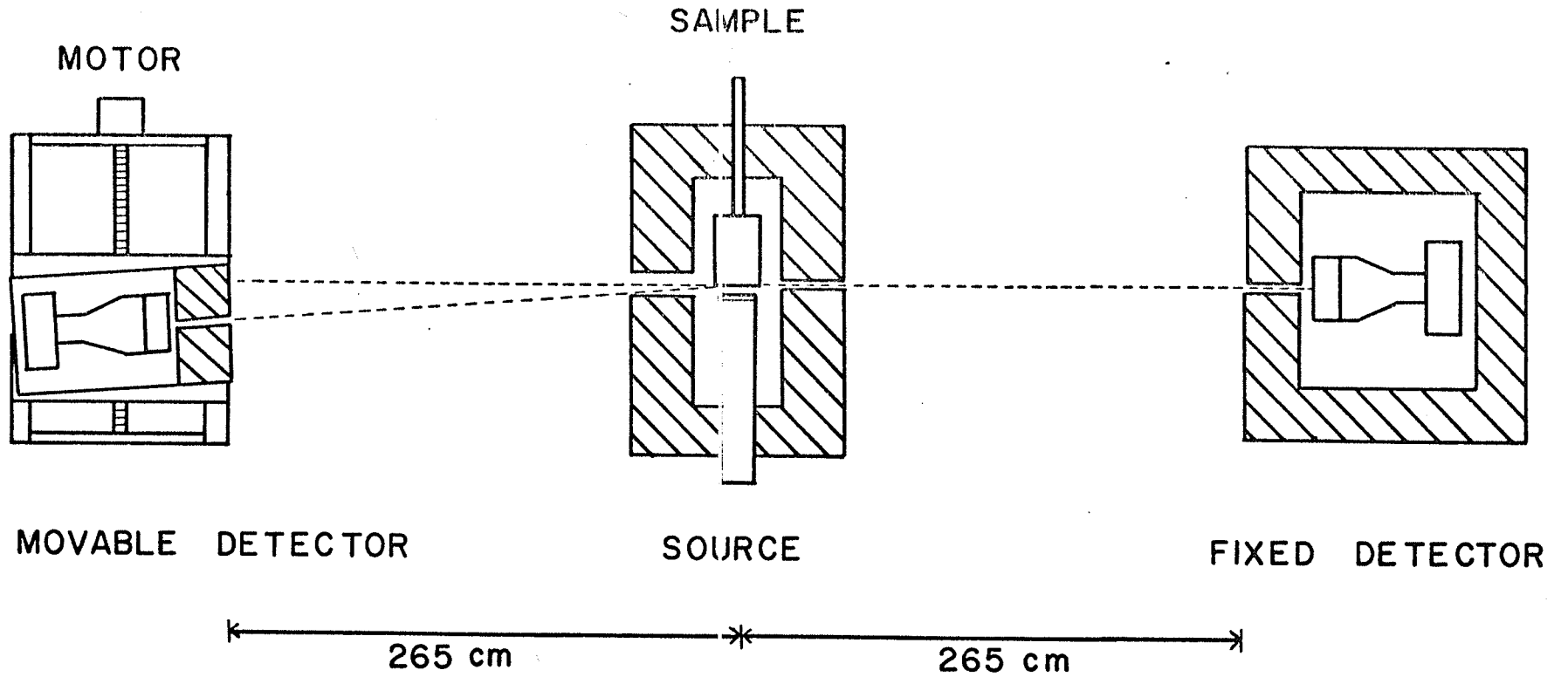
Magnetic Fields: L. A. Page and M. Heinberg 1956, G. Iaci et al. 1962, G. Fabri et al. 1964

Chemical Quenching: T. A. Pond 1952, S. Berko, A. J. Zuchelli 1956, D. A. L. Paul 1958, J. Lee and G. J. Celitans 1965, D. P. Kerr, A. M. Cooper and B. G. Hogg 1965.

From these studies certain insights into the various

Figure 1-1  
Angular Correlation Apparatus





mechanisms by which triplet positronium can have its lifetime shortened or quenched have been noted.

Foremost among the quenching processes would seem to be the "pick-off" process since in all condensed media this process is the basic one governing the triplet positronium lifetime. In the "pick-off" process due to the continual scattering of the positronium by the surrounding molecules, the positron in triplet positronium may annihilate with an electron from a molecule whose spin relative to the positron is anti-parallel. This process accounts for the long lived lifetime of positronium of about  $10^{-9}$  seconds in condensed medias rather than the lifetime of  $1.4 \times 10^{-7}$  seconds predicted for triplet positronium in free space.

The second quenching process we consider is that caused by an external magnetic field. In the ground state triplet positronium ( $3S_1$ ) is about  $8.4 \times 10^{-4}$  eV above the ground state of singlet positronium ( $1S_0$ ). When a constant magnetic field is applied the levels in triplet positronium are split and in the case of the  $m = 0$  levels in the singlet and triplet cases the energy separation between them becomes greater. This increases slightly the transition probability between them resulting in an increased probability of triplet to singlet conversion or quenching.

A third process considered possible in the quenching

of triplet positronium is that of paramagnetic quenching. In this case the quenching occurs not only from the action of an external field but also from the fields of paramagnetic admixtures in the sample. Ore (1949) has shown however that even for a gas as paramagnetic as oxygen the spin flip through magnetic interaction would take of the order of  $10^{-5}$  seconds, which is two orders of magnitude less than the case for spontaneous three photon decay. Therefore it would seem unlikely that this process is of any consequence in the quenching of triplet positronium.

The final type of quenching may be considered under the heading of chemical quenching. Under this heading would be included the chemical reactions of addition, substitution, oxidation and the possibility of the formation of positronium compounds.

**In this work studies are conducted to ascertain the effect on positron lifetimes and intensities due to variation in temperature and change of phase for various substances. From information of this type some of the theories and models currently proposed to explain positron decay processes can be tested. Also from studies of this type information is gained as to the usefulness of positrons as probes in studying various materials.**

## CHAPTER II

### INSTRUMENTATION

#### 2.1 Introduction

The measurement of time intervals between various nuclear or atomic events has been a study important to physics for many years. Originally studies of this type were conducted using simple coincidence elements which gave an output when two input pulses arrived at the coincidence element within a given resolving time. Many circuits were designed to achieve this end and almost all worked satisfactorily. The next development in this field was the addition of pulse amplitude selection to each of the two input sides to allow identification of the events under study. This was originally done by placing pulse height analyzers prior to the coincidence element in the circuit. This method however had the defect that the slow response of the pulse height analyzers severely limited the resolution obtainable by the coincidence circuit. This problem was overcome through the use of the fast-slow principle first described by Bell and Petch in 1949. In their method the fast coincidence was obtained for pulses unselected as to amplitude while the pulse height analysis was performed on pulses unselected as to time. Then through the use of a slow coincidence circuit pulses which satisfied both criteria were identified.

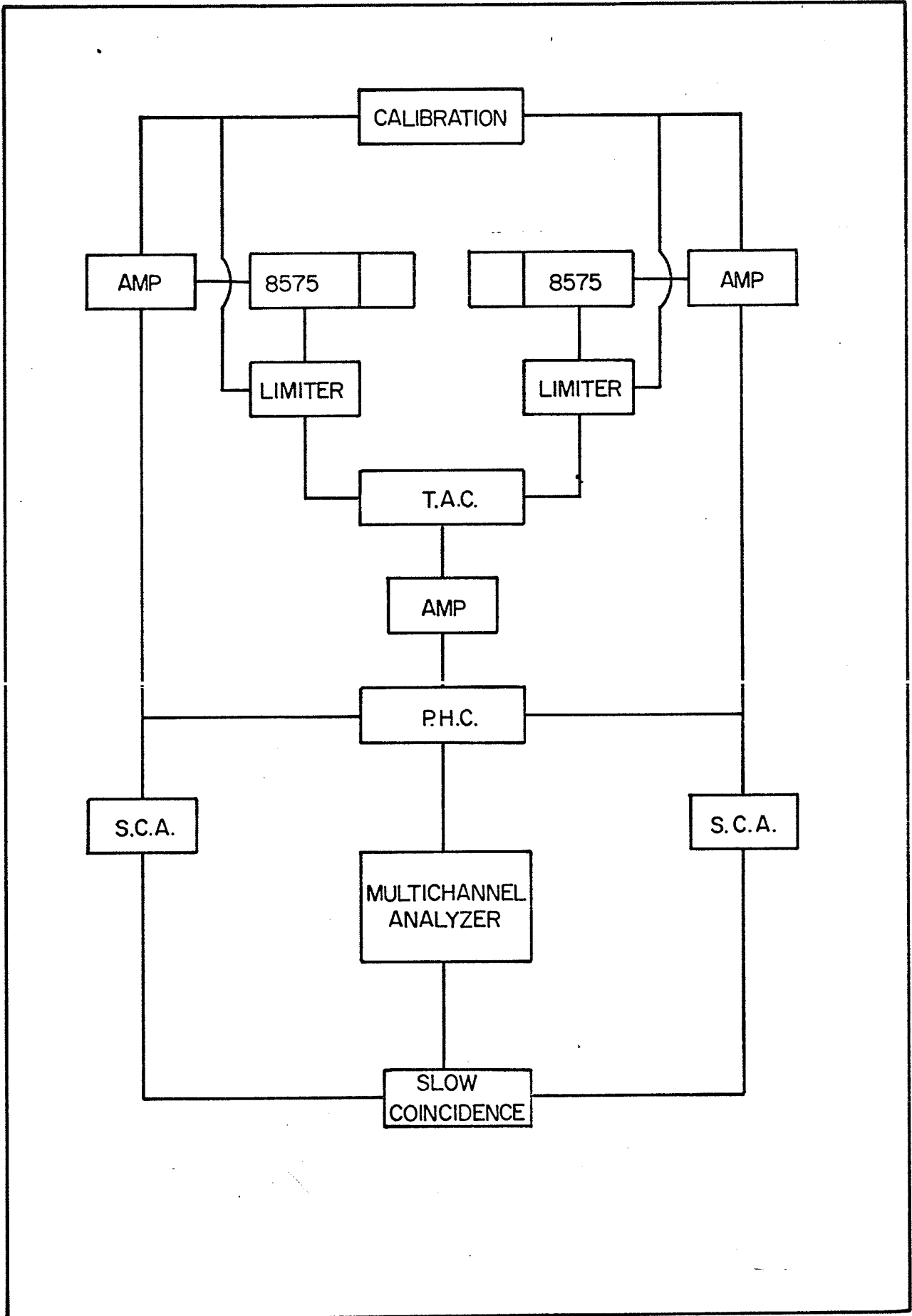
The fast coincidence circuit discussed thusfar only

gives information as to whether two events fall within a given time range, rather than giving the time distribution of the events. To achieve time distributions, experiments were originally performed by the placing of known delays between the two sides and counting for a given period of time at each different delay. This method was rather tedious but did achieve the desired effect of producing a time distribution of the events under consideration. Finally Time to Amplitude Converter (T.A.C.) circuits were introduced for use in the nsec region (Fraser and Milton 1953) which allowed time differences to be converted to voltage pulses which could then be displayed on multichannel analyzers. With the advent of this type of circuit we are at the level of present day time sorting systems. Although various other modifications have become standard and resolution obtainable has steadily improved the fast-slow system employing a T.A.C. has become standard for time measurements in the nsec region.

## 2.2 General Description of the System

A block diagram of the time sorting system used in this work is shown in Fig. 2-1. The system is conceptually the same as the fast-slow system described by Bell et al. (1952). Most of the actual circuitry was designed and built at the University of Manitoba. The system consists of three separate parts; the Fast side, the Slow side and the calibration section. Each of these various parts of the circuit will be described

Figure 2-1  
Block Diagram of the Time Sorting System



The Time Sorting System