ANGULAR CORRELATION OF ANNIHILATION RADIATION FROM POSITRONS ANNIHILATING IN SOME SIMPLE ALKANES

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ABSTRACT

The angular correlation of gamma rays from annihilating positron-electron pairs in liquid pentane and in solid and liquid butane has been studied. The angular correlation data have been converted to momentum distributions of the annihilating pairs. These momentum distributions are compared with theoretical calculations.

CHAPTER I

INTRODUCTION

The positron was first detected by Anderson in 1932 although it had been predicted by Dirac in 1930 from his relativistic wave equation. The positron has the same rest mass as the electron and has a charge +e where -e is the charge on an electron. An electron and a positron can annihilate with the emission of gamma photons.

Yang[1] has shown by means of charge conjugation invariance that a positron-electron pair with anti-parallel spins (singlet state) annihilates with the emission of an even number of photons while a positron-electron pair with parallel spins (triplet state) annihilates with the emission of an odd number of photons. One photon annihilation can only take place in the presence of an external field, and has an extremely low cross section even in the fields encountered in condensed matter. Two photon annihilation is the most probable annihilation process. Three photon annihilations have a cross section smaller by about two orders of magnitude. For higher numbers of gamma rays the cross section diminishes correspondingly.

When a positron enters condensed matter (liquid or solid) it may annihilate directly with an electron or it may capture an electron to form a bound state known as positronium. The positronium may be either in the singlet state or in the triplet state. The lifetime of singlet positronium is approximately 10^{-10} seconds and the lifetime of triplet positronium is approximately 10^{-7} seconds. In the time spectra of positron annihilations in condensed matter a third lifetime of approximately 10^{-9} seconds is observed. This is interpreted as being due to pickoff from the triplet state by atomic electrons whose spin states are singlet relative to the positron.

The mechanics of positronium formation in condensed matter was first discussed by Ore[2]. Like other charged particles, positrons lose energy when passing through matter, mainly through excitation of atoms and molecules. As the positron slows down to energies of a few tens of electron volts, positronium formation becomes possible. In order to form positronium, the positron must free an electron from a molecule of the surrounding matter. Therefore if the first ionization potential of a molecule is I the positron must have at least an energy of I - 6.8* eV in order to free an electron. On the other hand if the energy of the positron is greater than I, then positronium formation becomes very unlikely. Electron removal to the continuous spectrum becomes more probable than the formation of a discrete bound state. Moreover, if positronium does form it has kinetic energy greater than 6.8 electron volts and so it is exceedingly likely to dissociate

* 6.8 eV is the binding energy of positronium.

in subsequent collisions. Therefore positronium formation is a significant process only in the energy range from I - 6.8 eV to I. This range is known as the Ore Gap.

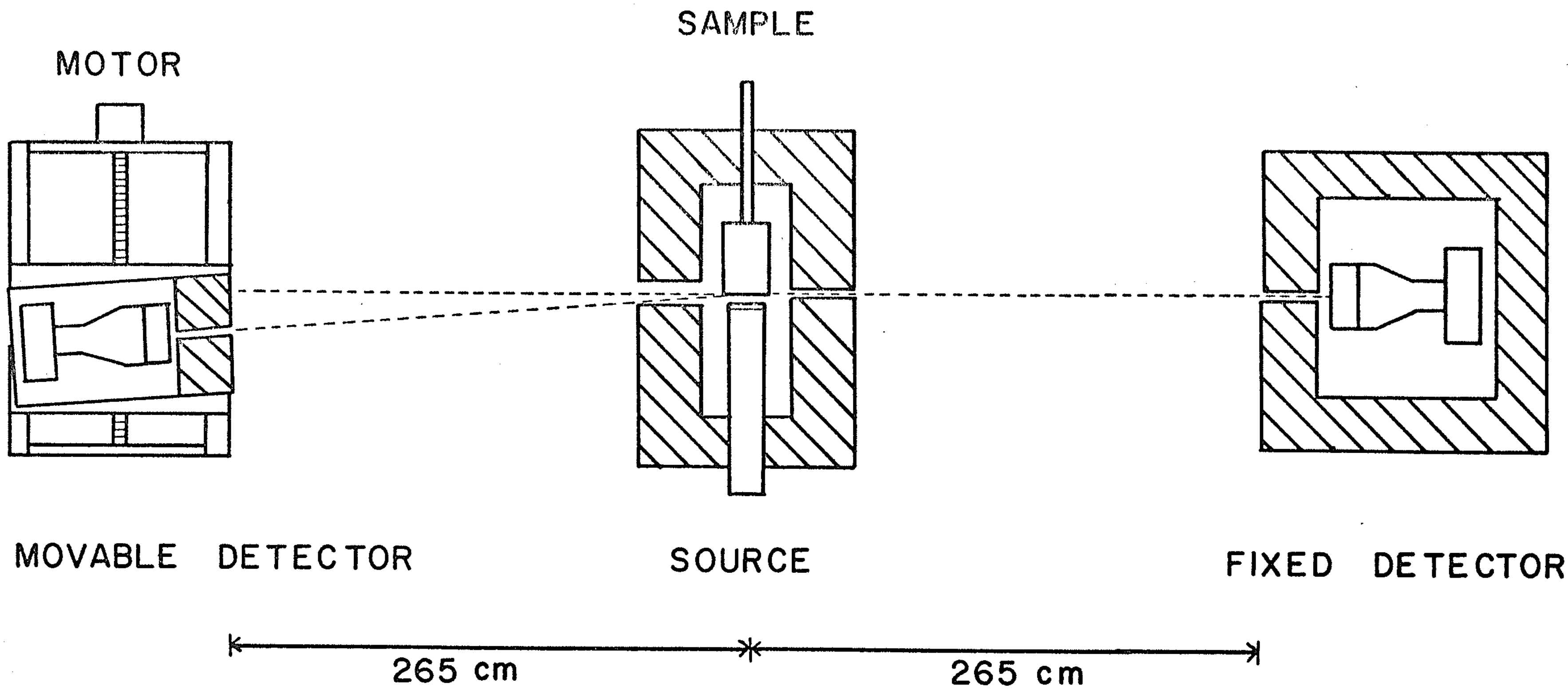
If a positron-electron pair at rest annihilates by two photon emission, in order to conserve momentum the two photons are emitted at an angle of 180° to each other, and in order to conserve energy each photon has an energy mc^2 where m is the rest mass of an electron. If the positron-electron pair is not at rest then the angle between the photons will differ from 180° and the energy of the photons will differ from mc^2. Assuming the momentum of the pair to be small compared to mc, the departure from co-linearity will be proportional to the component of momentum of the pair parallel to the bisector of the angle between the two photons. Therefore, by measuring the angular distribution of annihilation photons, one may determine the momentum distribution of annihilating positron-electron pairs.

In the present experiment, the angular distribution of annihilation photons is measured in one plane by means of a long slit apparatus. A schematic diagram of this apparatus is shown in Figure 1. Through certain mathematical steps to be described later, the momentum distribution of the positron-electron pairs can be determined from the angular distribution.

The original measurements of the angular correlation of annihilation photons were made by Beringer and Montgomery[3] in

Figure 1

Schematic Diagram of Angular Correlation Apparatus


SAMPLE
MOTOR
MOVABLE DETECTOR
SOURCE
FIXED DETECTOR
265 cm
265 cm

1942. Since then extensive work has been done by de Benedetti et al.[4] and by Stewart [5]. De Benedetti was the first to describe a method for obtaining momentum distributions from the angular distributions. D. Kerr[6,7] has since measured angular distributions in organic liquids. Chuang[8,9,10] has measured angular distributions in organic liquids with better statistics. He has also done theoretical calculations and obtained good agreement with experiment.

CHAPTER II

APPARATUS

A. Mechanical

The angular correlation apparatus consisted of a source and sample housing located halfway between two gamma ray detectors. The sample housing consisted of an insulating polystyrene box and two collimating slits. The gamma detectors consisted of NaI crystals with collimating slits. These components were mounted on two parallel 3" by 6" aluminium I-beams approximately 20 feet long.

One detector was fixed and the other movable. The fixed detector and its collimating slits were mounted on a brass plate 265 cm. from the source. The movable detector was mounted on a steel plate 265 cm. on the other side from the source. The steel plate was moved along a set of rails by means of a worm screw driven by a 600 ounce-ince "Slo-Syn" motor.

The collimating slits at the sample housing were made of 2" thick lead. The width of the slits were .15 cm. and 1.5 cm. facing the fixed and movable detectors respectively. The larger slit facing the movable detector was necessary in order that gamma rays from the sample could reach the detector for a whole range of positions.

The collimating slits of the detectors were made from 3" thick lead blocks. This was sufficient to cut down the intensity of the .511 MeV and 1.27 MeV gamma rays to less than .2% of the incident value. The width of the collimating slits could be changed by inserting metal shims of appropriate thickness. During the present experiment the slits were .15 cm. wide.

An aluminium beam was used to keep the collimating slit of the movable detector aligned towards the sample as the detector moved through different positions.

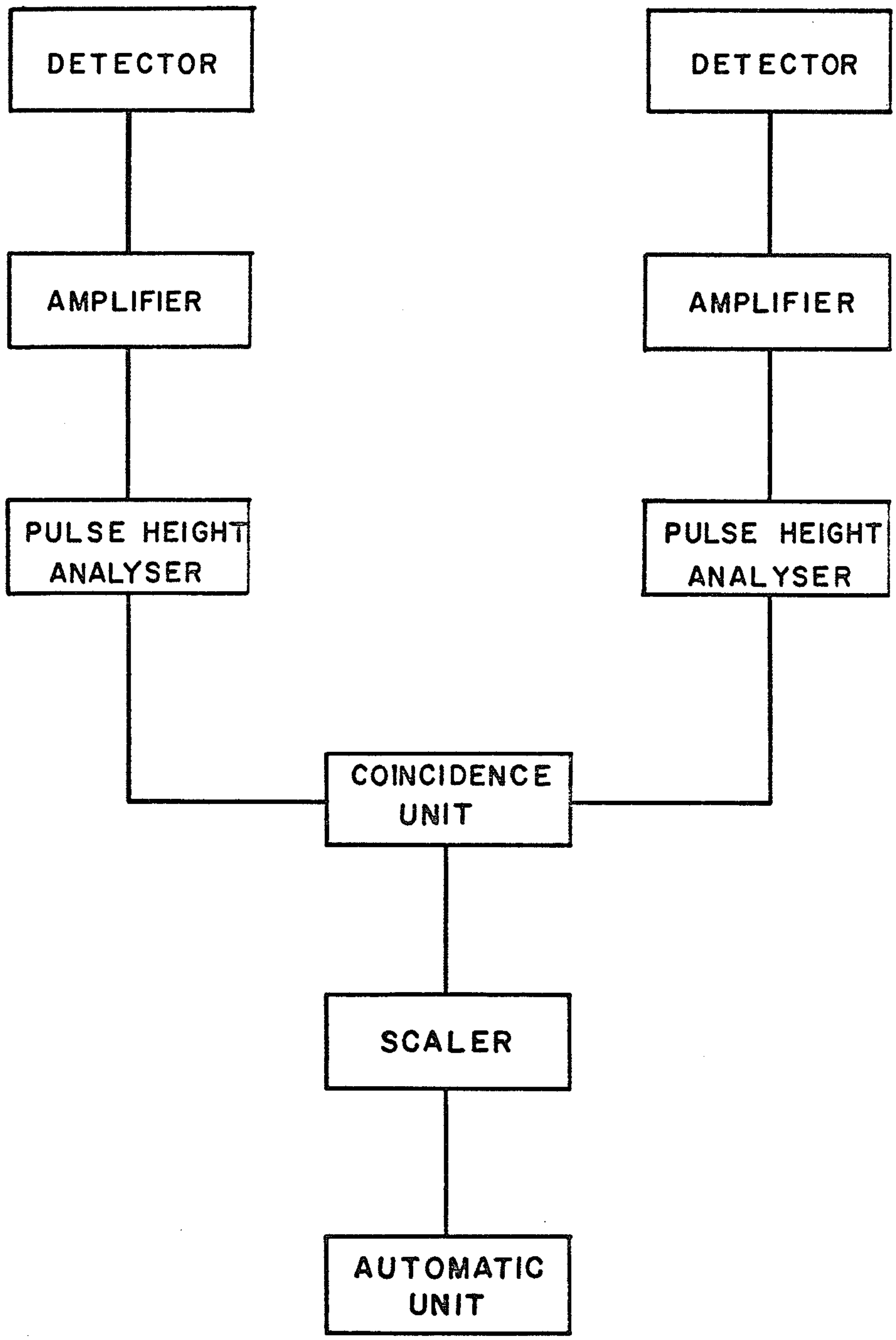
B. Electronic

The gamma detectors were Integral Assembly model 16MB4/A-X. The scintillator in each assembly was a 4" diameter, 1" thick NaI(Tl) crystal. This was mounted on a 5018 HB photomultiplier. A positive potential of 1050 volts was applied to the photomultipliers by a Hamner N401 high voltage supply. Cathode followers in the detector heads were used to feed negative pulses (approximately 1 volt) to two simple transistorized amplifiers. The amplifiers increased the pulse height about five times and shortened the pulse length to about one microsecond.

From the amplifiers the pulses were sent to a pair of single channel analyzers. By means of a Tektronix 541A oscilloscope the single channel analyzers were set to select gamma rays between .1 and .6 MeV. This allowed through the

Figure 2

Block Diagram of Electronics


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

Doppler shifted .511 MeV spectrum from the annihilations but blocked out the low energy noise and the 1.27 MeV γ-rays from the source. The fast rising, narrow pulses from the single channel analyzers were sent to a single coincidence unit.

To determine the resolution time of the coincidence unit, a random source was provided for each counter. The individual counting rates N_1 and N_2, and the chance coincidence rate c.c. were measured. Then the resolution time T was calculated from the relation

$$c.c. = 2TN_1N_2$$

This procedure was carried out before the start of each run. The resolution time was found to be between 100 and 200 nanoseconds.

The number of coincidences was recorded by a Technical Measurement Corporation SG-3A scaler. When a certain prescribed number of counts (usually 400 or 1000) had been accumulated in the scaler, the movable detector was automatically moved to its next position. The electronics used to achieve this automatic movement has been described elsewhere (Ref. 7). The scaler stops counting while the detector is being driven to its new position and starts again when the detector stops. At a predetermined position the direction of motion of the detector is reversed by a micro switch.

A Simplex ET-100 interval timer was used to record the time spent at each position to accumulate the required number of counts. All the electronics was powered by a Sorenson model 2000 S A.C. voltage regulator.

C. <u>Sample and Source Arrangements</u>

The sample cell was constructed of 1/32" aluminum. A window of .001" aluminum across one end of the cell permitted positrons to enter the sample. It was necessary to have the window this thick in order that the sample cell should be capable of holding a vacuum better than 10^{-3} torr. The volume of the sample cell was approximately 7 ml.

During the experiment, the sample cell was positioned so that only annihilations in a region just inside the window were recorded. Due to this and the fact that very few positrons penetrate through the sample and reach the walls of the cell, no correction was needed for annihilations in the aluminum.

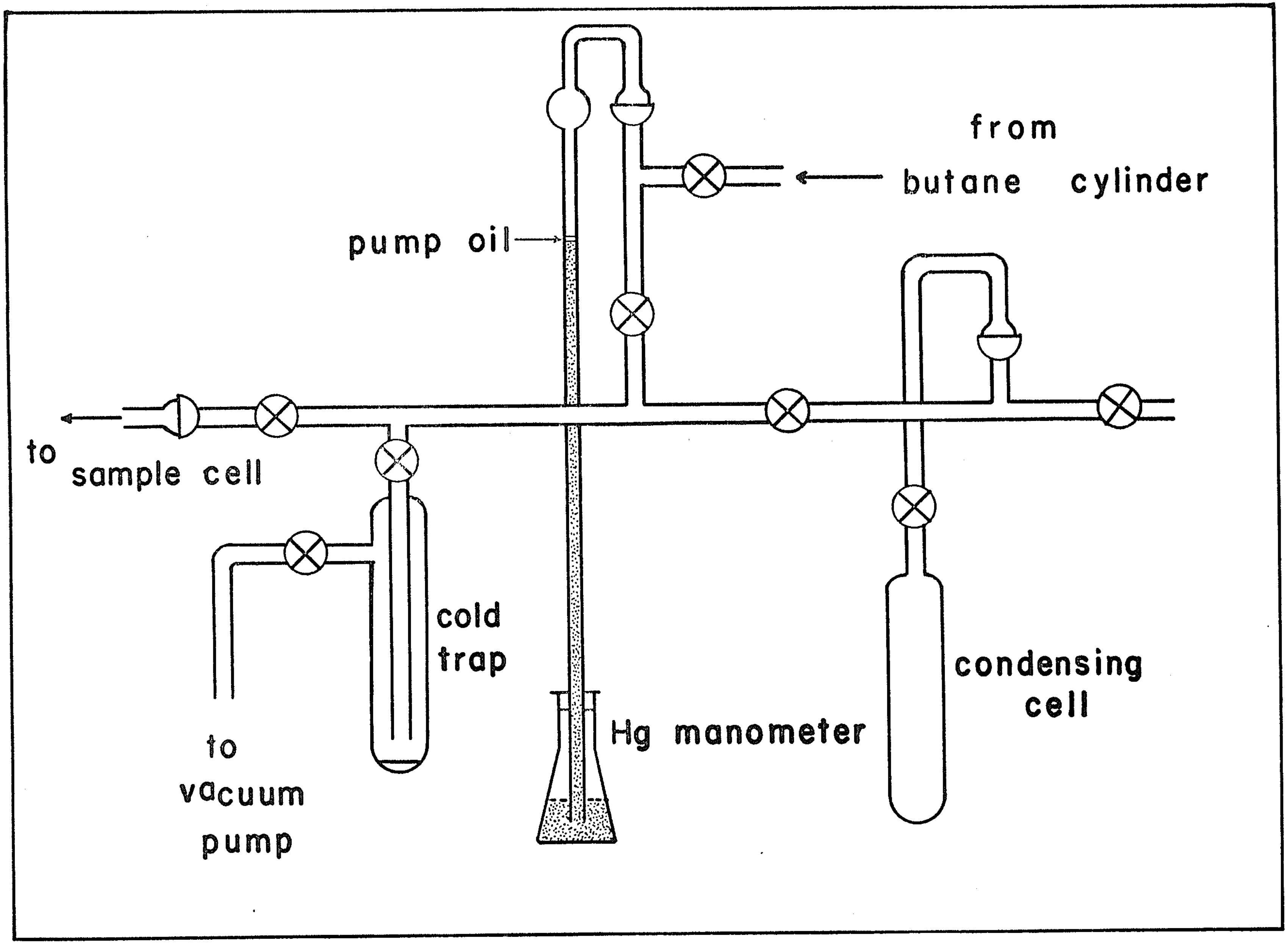
The Butane used in the experiment was research grade obtained from Matheson of Canada, Limited, Whitby, Ontario. This had a purity of 99.96 mol %.

To prevent contamination of the sample the Butane was transferred to the sample cell, by means of the system shown in Figure 3. This apparatus used high vacuum stopcocks and ground glass joints sealed with high grade Apiezon grease. A thin layer of vacuum pump oil on the mercury in the manometer prevented evaporation of mercury into the system.

To transfer the Butane, the system was pumped down to

Figure 3

Distillation Apparatus


from
butane cylinder
pump oil
to sample cell
cold trap
to vacuum pump
Hg manometer
condensing cell

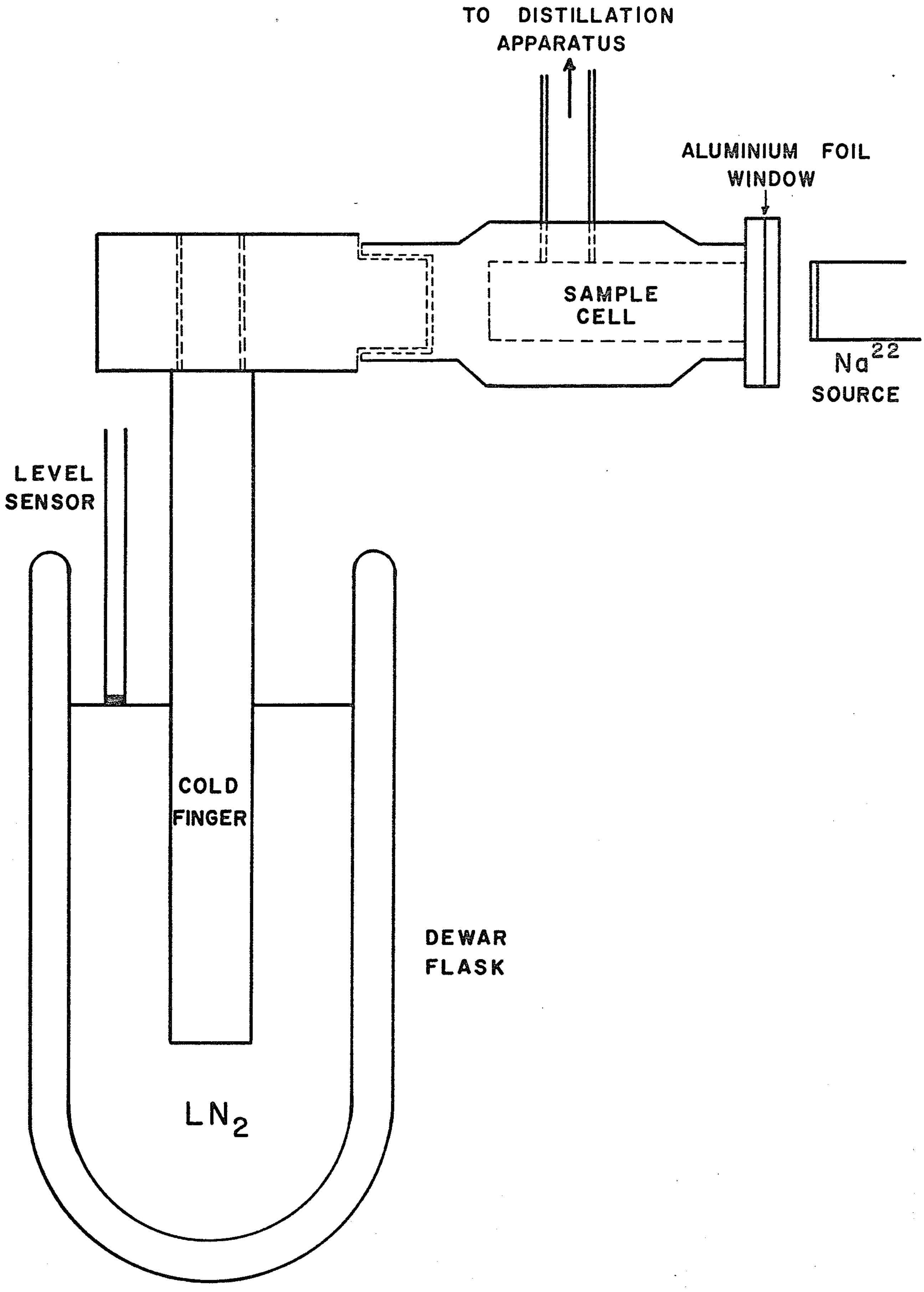
a pressure of less than 10^{-3} torr. The Butane was then condensed into the condensing cell which was cooled by liquid nitrogen. The sample cell was then cooled by liquid nitrogen and the condensing cell was allowed to warm to room temperature thereby causing the Butane to transfer from the condensing cell to the sample cell.

To remove the air dissolved in the Pentane it was subjected to the standard freeze-thaw technique. The same sample cell was used, however it was disconnected from the distillation apparatus. The Pentane was poured into the sample cell and the sample cell connected to a vacuum pump. After the sample was degassed the cell was clamped off and disconnected from the vacuum pump. The sample cell was then placed in position in the angular correlation apparatus.

To keep the Butane at the correct temperature a cold finger arrangement was utilized. A cross section is shown in Figure 4. To maintain the level of the liquid nitrogen in the dewar and thus keep the temperature within prescribed limits, an electronically controlled valve was employed. A circuit diagram and description of the operation of the circuit are given in Ref. 10. Instead of using an electric air pump as in the preceding reference, the boil-off from the liquid nitrogen in the storage dewar was used to force the liquid nitrogen through the transfer tube. With this arrangement the transfer time was between 5 and 10 minutes and the temperature variation

Figure 4

Sample Cell and Cold Finger Arrangement


TO DISTILLATION
APPARATUS
ALUMINIUM FOIL
WINDOW
SAMPLE
CELL
Na22
SOURCE
LEVEL
SENSOR
COLD
FINGER
DEWAR
FLASK
LN2

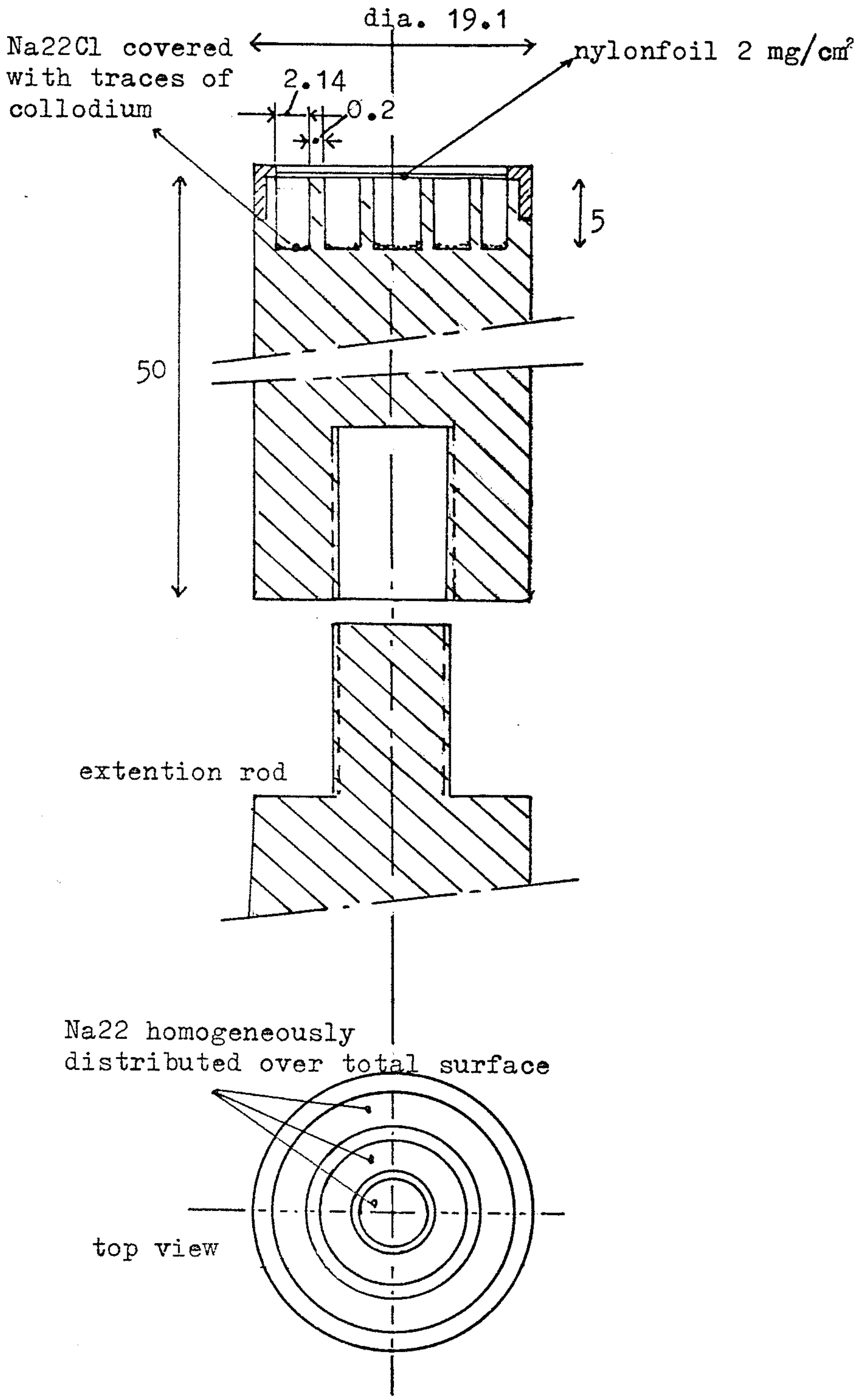
was about plus or minus 5 degrees. Since the temperature was not deemed to be critical except very close to the melting point, this was considered to be satisfactory temperature control.

To measure the temperature in the sample cell a copper-constantan thermocouple was used. One junction was cemented to the sample cell with epoxy glue. The other junction was kept at 0°C by immersion in an ice and water mixture. The potential difference between thermocouple junctions was measured by means of a potentiometer and the temperature was determined with the aid of a calibration table.

The source of positrons used in these experiments was a $Na^{22}Cl$ source purchased from N. V. Philips-Duphar Isotopes Division. The source consisted of $Na^{22}Cl$ deposited in concentric grooves milled in the end of a Perspex rod 19.1 mm. in diameter as shown in Figure 5. The end was then covered with a round nylonfoil disk of thickness 2 mg./cm^2. The grooves tended to direct the beam of positrons along the axis of the rod. The perspex rod was then connected to a lucite rod of the same diameter to allow manipulation in the source castle. Originally the source was 20 millicuries, however by the time of these experiments it was about 7 or 8 millicuries.

Figure 5

Positron Source



Measures in mm.

CHAPTER III

DATA ACCUMULATION AND ANALYSIS

To accumulate data, the reversing microswitches were set at convenient positions and the movable detector was set at one of the microswitches. The movable detector was then run back and forth between the reversing switches taking 400 counts at each point. This was continued until approximately 10,000 counts had been accumulated at each point. The points were .907 milliradians apart.

To interpret the count rate $c(\theta)$ we follow de Benedetti[4] and the more rigorous treatment by Stewart[5]. Take $\rho(\vec{p})$ to be the density in momentum space of the annihilating pairs. Choose a set of Cartesian axes such that the x-axis lies along the line joining the fixed detector to the sample cell, the y-axis lies along the axis of the detector slit, and the z-axis is perpendicular to the x and y axes. Neglecting the number of annihilating pairs having a y-component of momentum large enough that the resulting gamma rays would subtend an angle larger than that subtended by the slit , then the coincidence count rate is given by

$$c(p_z) = A\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\rho\,(p_x,\,p_y,\,p_z)\,dp_x\,dp_y$$

where $p_z = mc\theta$ as long as $p_z \ll mc$ and A is some constant depending on detector configuration and efficiency.

If the variables of integration are changed to

$$P = \sqrt{p_x^2 + p_y^2} \text{ and } \phi = \tan^{-1}(p_y/p_z)$$

and it is assumed that $\rho(\vec{p})$ is isotropic, then

$$c(p_z) = 2\pi A \int_0^\infty \rho(P, p_z)\, PdP$$

To evaluate this integral, the substitution

$$p^2 = P^2 + p_z^2$$

is made. Then

$$c(p_z) = 2\pi A \int_{p_z}^\infty \rho(p)\, pdp$$

This expression is then differentiated with respect to p_z, yielding

$$\frac{dc(p_z)}{dp_z} = -2\pi A\, p_z\, \rho(p_z)$$

It follows that

$$\rho(p_z) = -\frac{1}{2\pi A p_z} \frac{dc(p_z)}{dp_z}$$

This $\rho(p_z)$ is the density in momentum space of annihilating pairs having momentum of magnitude p_z.

Now since $p_z = mc\theta$

$$\rho(p_z) = -\frac{1}{2\pi A m^2 c^2} \frac{1}{\theta} \frac{dc(\theta)}{d\theta}$$

$$= -B \cdot \frac{1}{\theta} \frac{dc(\theta)}{d\theta}$$

where B is some positive constant.

The area of a sphere in momentum space of radius p_z is $4\pi p_z^2$. Therefore $N(p_z)$, the number of annihilating pairs having momentum of magnitude p_z is given by

$$N(p_z) = -4\pi B m^2 c^2 \theta \frac{dc(\theta)}{d\theta}$$

In the following sections the data will be presented in the form of N(p) or $c(\theta)$. Since the constant B is not known the term $\frac{dc(\theta)}{d\theta}$ is simply multiplied by a normalizing factor. In these experiments the curves are normalized to a peak of 10.

The actual analysis of the data was done by a computer program. The data was accumulated in the form of the time intervals required to accumulate 400 counts. These times were recorded by the Simplex ET-100 timer and then transferred to punch cards. These cards together with other pertinent data were fed into the computer. The program first corrected the times for source decay and calculated the count rate $c(\theta)$ for each point. Next the program calculated the derivative at each point by means of a five point least squares fit to a parabola. For the 2nd and n-2th points a three point least squares fit to a parabola was used and for the end points a straight difference was used. The program then calculated N(p) and $\rho(p)$ and the error in N(p). Finally $c(\theta)$, the error in $c(\theta)$, $c^1(\theta)$, $\rho(p)$, N(p) and the error in N(p) were outputed for each point in tabular form. The $c(\theta)$ and N(p) curves are shown in Figures 7-12. The background was determined by counting with no sample cell in place. The resulting distribution was almost flat with a peak counting rate of less than 1 count per minute. The chance coincidence rate was well less than 1 count per minute.

The distribution was also measured with the sample cell in place, but no sample in the cell. This resulted in the inverted parabola type distribution shown in Figure 6. This agrees very well with other results for aluminium(Refs. 5, 11). This is interpreted as being annihilation in the window and walls of the sample cell. This distribution cannot be just subtracted from the experimental curves since when the sample is in the cell very few positrons penetrate far enough to annihilate in the walls and when samples were run care was taken to look at an area behind the window.

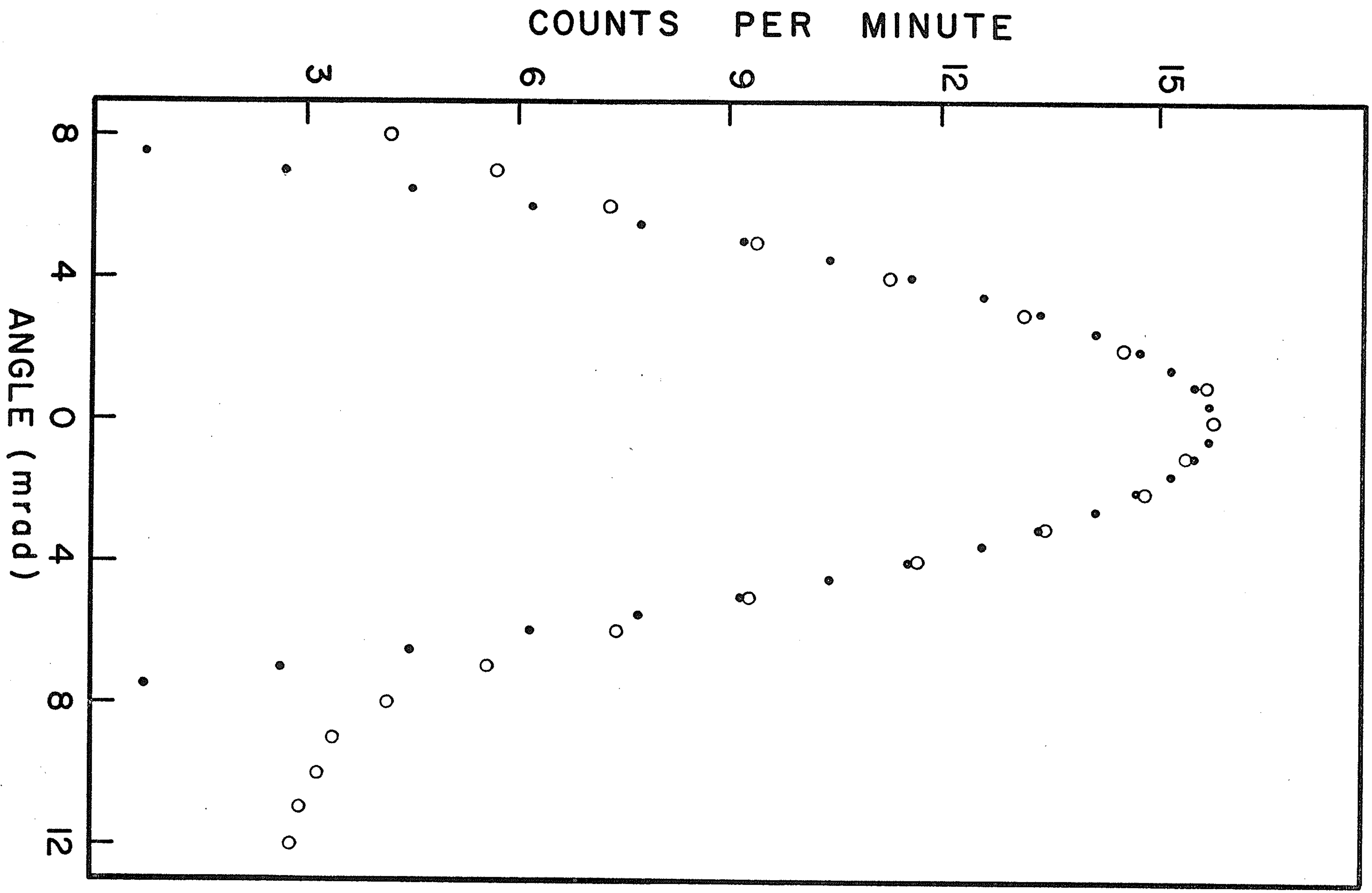
A computer program was used to subtract different percentages of this background from the experimental curves and plot the resulting distributions by means of a Calcomp 585 plotter. It did not make a significant change in the distributions.

Figure 6

Aluminium Background Angular Distribution

o ---- Experimental points

● ---- Parabola empirically fitted to the experimental points at the peak


COUNTS PER MINUTE
3
6
9
12
15
8
4
0
4
8
12
ANGLE (mrad)

CHAPTER IV

THEORETICAL CALCULATIONS

The momentum space wave function for an annihilating positron-electron pair has the form

$$\chi(p) = (2\pi)^{-3/2} \int e^{-i\vec{p}\cdot\vec{r}} \psi_-(r) \psi_+(r)\, dr$$

now N(p) is given by

$$N(p) = \int_0^{2\pi} \int_{-\pi/2}^{\pi/2} \chi^*(p)\, \chi(p)\, p^2 \sin\theta \; d\theta\, d\phi$$

$$= 2\pi \int_{-\pi/2}^{\pi/2} \chi^*(p)\, \chi(p)\, p^2 \sin\theta \; d\theta$$

As described by Chuang[8,9,10], knowing the positron wave function, one may use the hybrid carbon orbital wave functions together with the hydrogen 1s orbital wave function to calculate $\chi(p)$ and thence N(p) for the C-H and C-C bonds in a simple alkane.

The positron wave function is obtained by numerically solving the Schrödinger equation in a Wigner-Seitz cell. The atomic potential used for the hydrogen atom was the standard analytic potential[12]. The atomic potential for the carbon atom was the numerical one determined by Herman and Skillman[13] through a self-consistent field method.

Having determined the momentum distributions for the C-C and C-H bonds, the momentum distribution for a given alkane can be predicted under certain assumptions.

One assumes that only electrons from the C-H and C-C bonds annihilate with the positrons and that annihilation is equally probable for an electron from either bond. One then calculates a weighted average of the two distributions weighting N(C-C) and N(C-H) according to the relative number of electrons in each type of bond. For Butane and Pentane this gives the following formulae.

$$N(\text{butane}) = 6/26\ N(C\text{-}C) + 20/26\ N(C\text{-}H)$$

$$N(\text{pentane}) = 8/32\ N(C\text{-}C) + 24/32\ N(C\text{-}H)$$

The theoretical momentum distributions are shown as solid curves in Figures 10-12.

Figure 7

Angular Distribution for Liquid Pentane

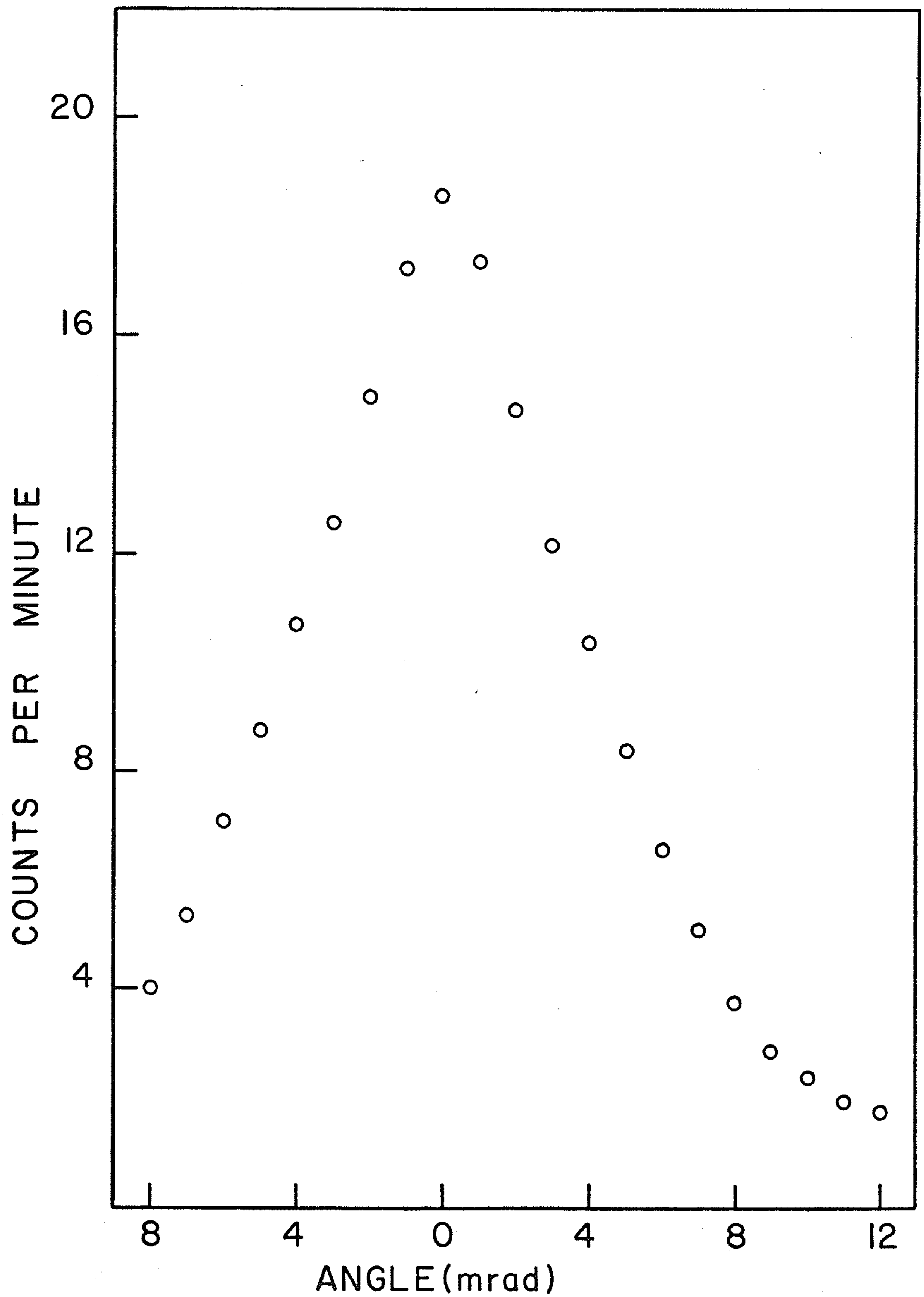

COUNTS PER MINUTE
20
16
12
8
4
8
4
0
4
8
12
ANGLE(mrad)

Figure 8

Angular Distribution for Solid Butane

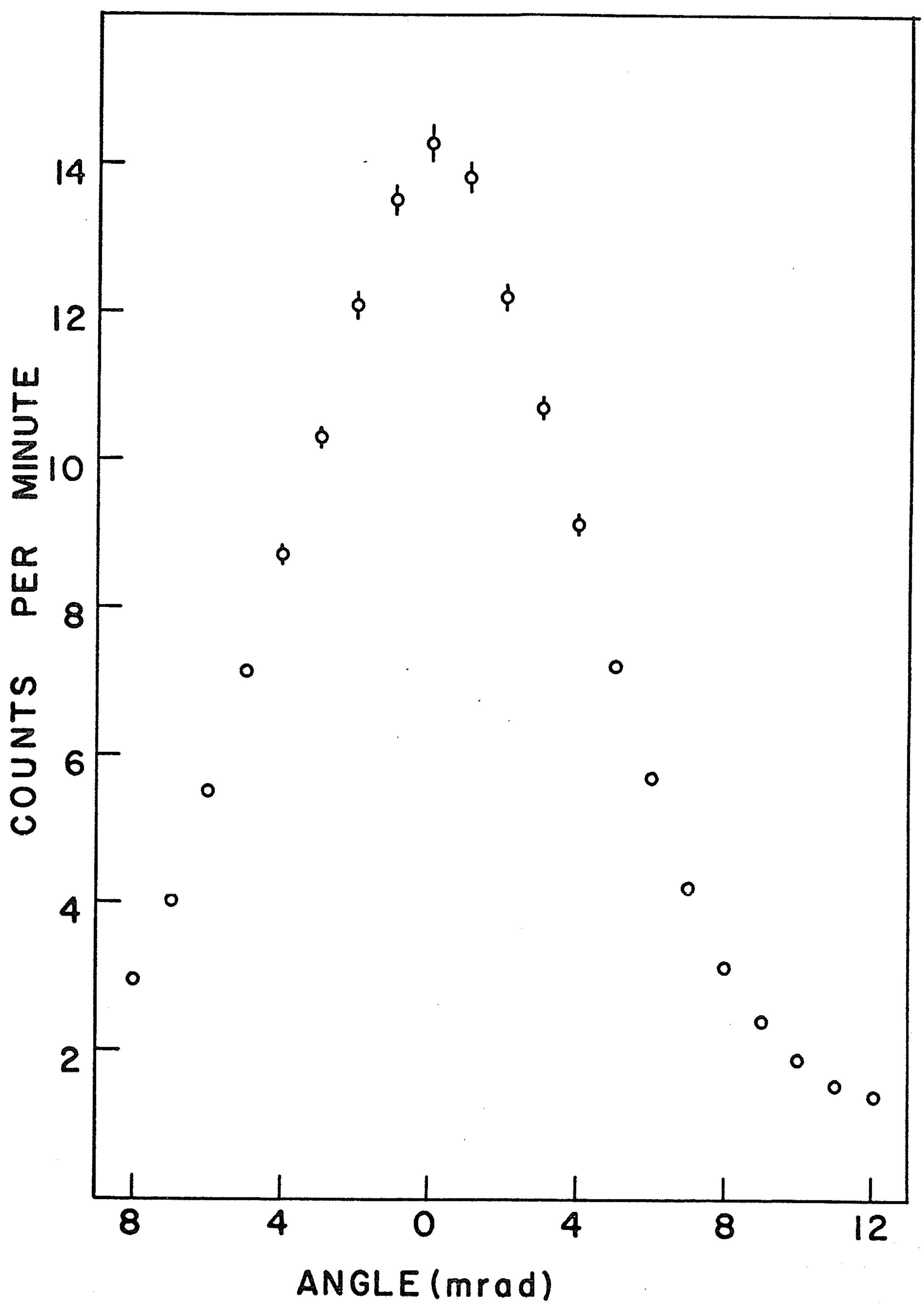
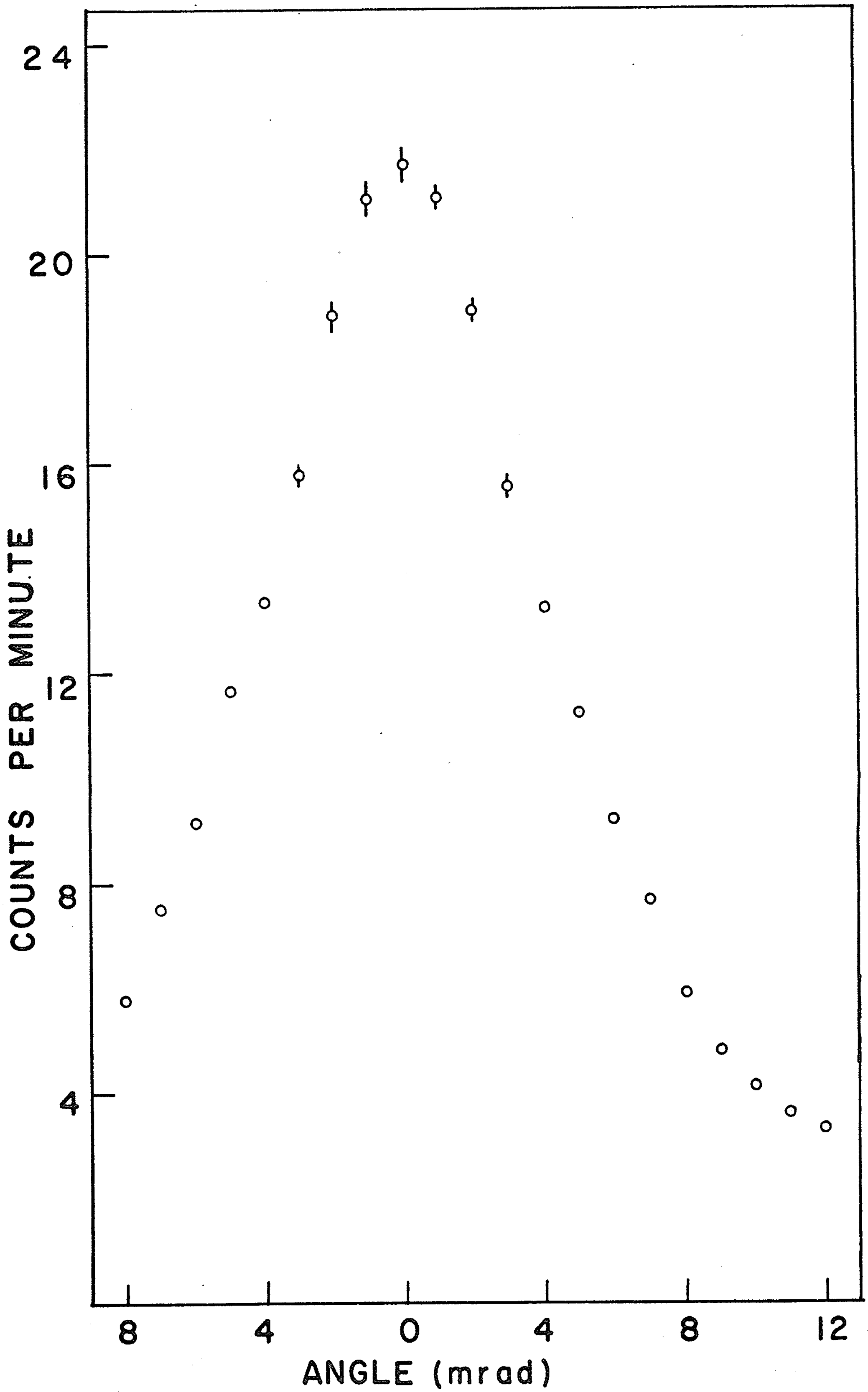

COUNTS PER MINUTE
2
4
6
8
10
12
14
8
4
0
4
8
12
ANGLE (mrad)

Figure 9

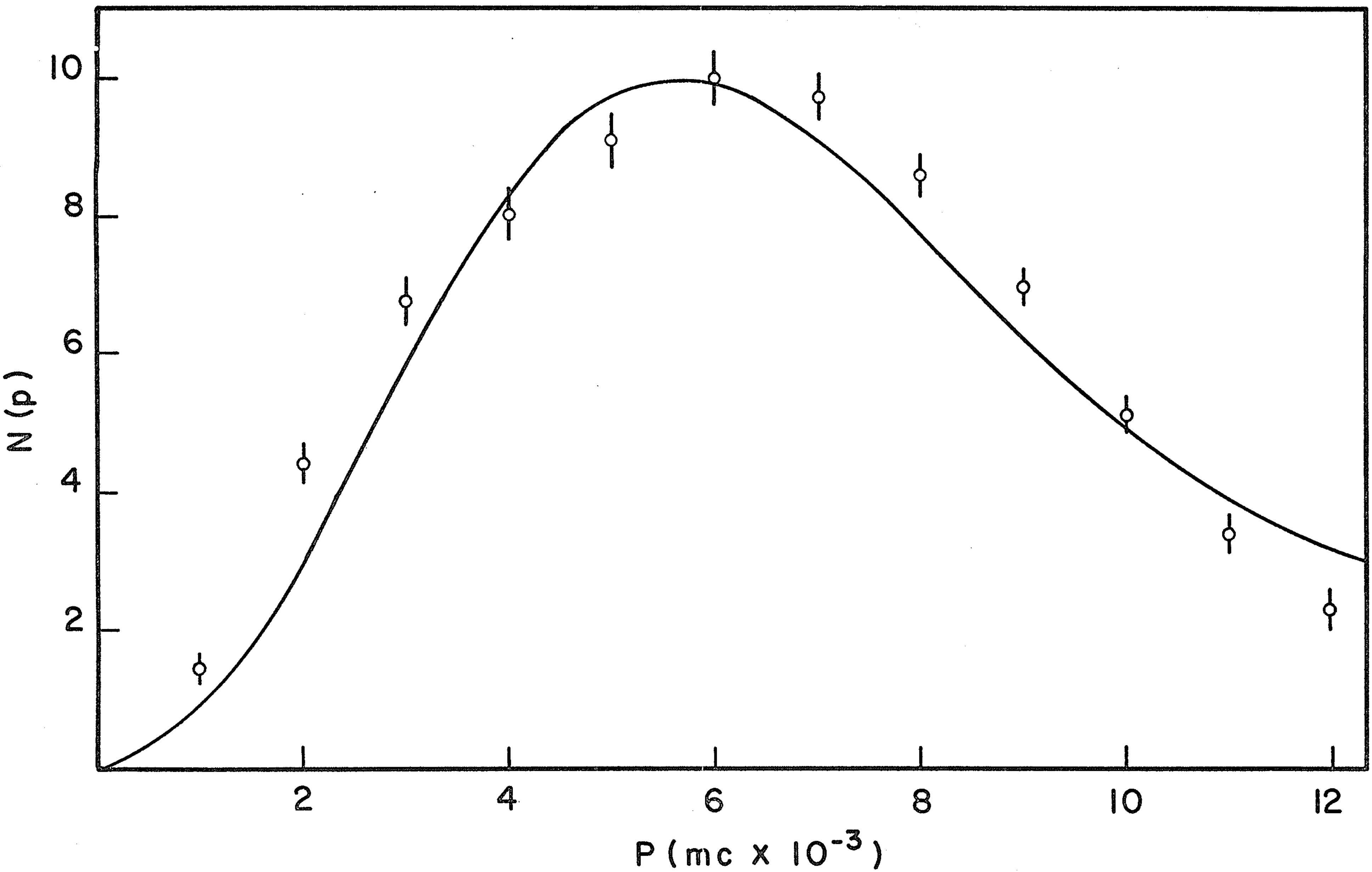
Angular Distribution for Liquid Butane



COUNTS PER MINUTE
24
20
16
12
8
4
8
4
0
4
8
12
ANGLE (mrad)

Figure 10

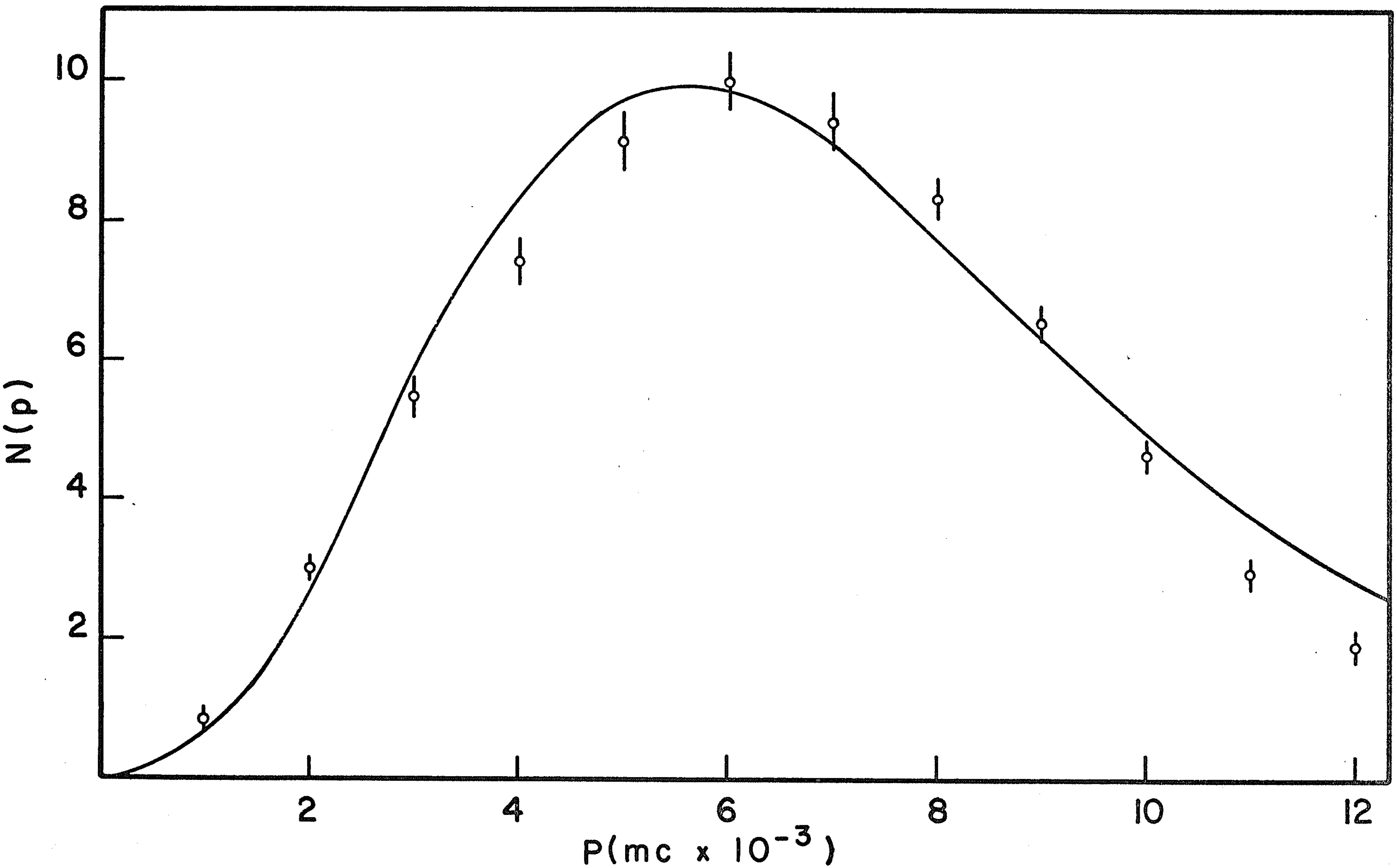
Momentum Distribution for Liquid Pentane



10
8
6
4
2
N (p)
2
4
6
8
10
12
P (mc x 10^{-3})

Figure 11

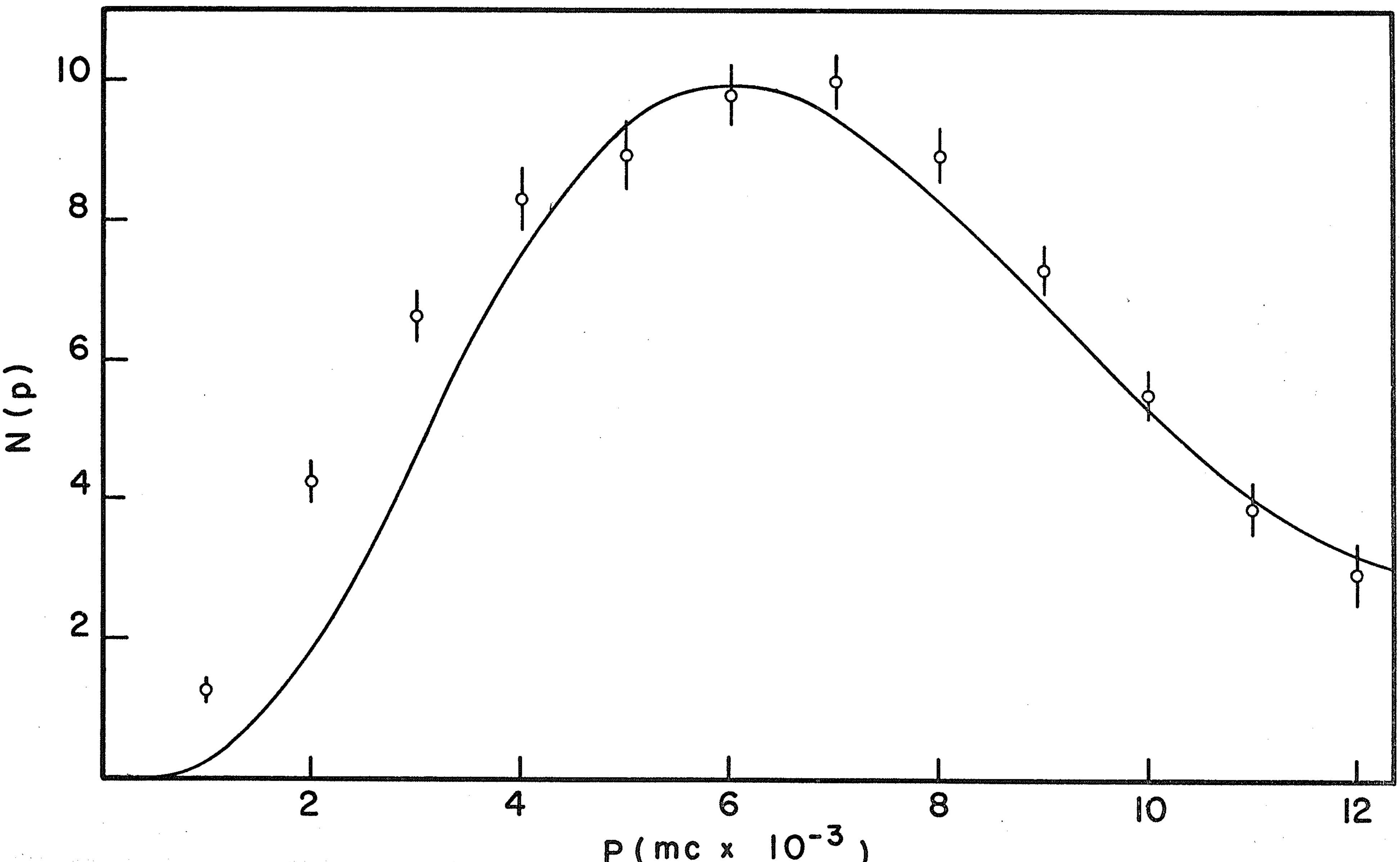
Momentum Distribution for Solid Butane



N(p)
10
8
6
4
2
2
4
6
8
10
12
P(mc x 10^{-3})

Figure 12

Momentum Distribution for Liquid Butane


N (p)
10
8
6
4
2
2
4
6
8
10
12
P (mc x 10^-3)

CHAPTER V

CONCLUSIONS

The theoretical and experimental curves do not quite coincide. As can be seen the theoretical fits need further work. A possible modification would be to include the contribution from the carbon 1s electrons. This would raise the high momentum end of the curve and result in better agreement with the experimental curve.

From a purely statistical argument three times as much triplet positronium should be formed as singlet positronium. Therefore if I_2 is the intensity of pickoff annihilations from triplet positronium as determined by lifetime measurements,[15] then I_L the intensity of singlet annihilations as determined from the momentum distributions should be equal to $I_2/3$. The following table shows these values.

	I_L (%)	$I_2/3$ (%)
Liqud Pentane	9	10
Solid Butane	4	4
Liquid Butane	6	6

The close agreement between I_L and $I_2/3$ is strong evidence that the statistical distribution of occupied states is in fact the distribution realized experimentally.

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