GAMMA RAY SPECTROSCOPY OF TIN - 113 AND OF ISOTOPES PRODUCED BY CYCLOTRON IRRADIATION OF COPPER

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A lithium drifted germanium detector and a NaI(Tl) sum coincidence spectrometer have been used in the study of the decay schemes of Sn^{113}, Zn^{62}, Cu^{62}, and Cu^{61}. A method is described for reducing the amount of positron annihilation radiation in the spectrum of a positron emitter. An ion exchange column was used for separating zinc and copper.

An upper limit of 0.04% has been set on the 648 keV cross over transition in In^{113}. The end-point energy of the inner bremsstrahlung radiation following the decay of Sn^{113} to the 393 keV level in In^{113} is found to be 680 $\pm$ 10 keV, giving a decay energy of 1.10 MeV.

The half-life of Zn^{62} has been found to be 9.3 $\pm$ 0.2 hours. Four new transitions in the decay have

been found at 303, 345, 549, and 640 keV. The energies and intensities of the others have been accurately established using the germanium detector.

The 1170 and 880 keV transitions in the decay of Cu^{62} have been confirmed.

The gamma rays in the decay of Cu^{61} have been studied and their energies and intensities measured.

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Abstract

A lithium drifted germanium detector and a (NaI(Tl) sum coincidence spectrometer have been used in the study of the decay schemes of Sn^{113}, Zn^{62}, Cu^{62}, and Cu^{61}. A method is described for reducing the amount of positron annihilation radiation in the spectrum of a positron emitter. An ion exchange column was used for separating zinc and copper.

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The gamma rays in the decay of Cu^{61} have been studied and their energies and intensities measured.

Chapter 1

INTRODUCTION

The atomic nucleus is a complex structure, and the theory describing it is in a rather incomplete state. Because any present theory that would describe all aspects of the nucleus would have too many parameters to be easily visualized by the human mind, various models have been proposed to describe different sets of properties of the nucleus. These are the optical model for scattering theory, and the shell model and the collective model for description of energy levels, to mention a few.

The shell model, or single particle model in particular, is applied to decay schemes. In this model the spin and parity of a nuclear state are given by a single nucleon, the rest of the nucleus being thought of as a closed system. Since a gamma ray represents an electromagnetic transition between two energy levels in a nucleus can then be determined from a knowledge of the gamma spectrum emitted. It is hoped that when enough knowledge about the nucleus has been gathered it will be possible to set up a comprehensive theory describing it.

In the study of decay schemes an important experimental result is the decay energy, from ground state to ground state. For electron capture decays, this energy can sometimes be measured through the inner bremsstrahlung spectrum, and this was the prime purpose of the study of Sn^{113} .

THEORY ON INNER BREMSSTRAHLUNG

There exists a probability that a nucleus $^{A}_{Z}X$ having another nucleus $^{A}_{Z-1}Y$ with lower total energy as its neighbor, will decay to this state. If the total decay energy available is greater than 1.022 MeV, then both positron emission and electron capture transitions are possible modes of decay. This decay will occur with a characteristic half-life which is given by the sum of the individual decay probabilities of positron emission and electron capture. In the latter, an orbital electron is captured by the nucleus which then emits a neutrino. The probability of this capture is roughly proportional to the amount of overlap of the wave functions of the electron and the nucleus. For this reason K-electron capture predominates over L-capture transitions whenever this is energetically possible. Since positron emission involves the creation of a positron emission involves the creation of a positron, it is possible that electron capture will be energetically possible even though positron emission is not. The ratio of the probability of positron emission to the probability of electron capture for a nucleus of given atomic number is given by Bouchez and Depommier (Bouchez and Depommier, 1960).

Accompanying K capture there is a weak

continuum of high energy gamma radiation called "Inner bremsstrahlung". Theoretical calculations have been done on this (Morrison and Schiff, 1940) using second order perturbation theory, assuming either Fermi or Gamow - Teller couplings for the weak interaction causing the decay of the nucleus. Their result for the distribution of intensity with energy of this inner bremsstrahlung is of the form

$$(N(k)/k)^{1/2} = \text{const}(w-k)$$

where k is the energy and N(k) is the number of photons at this energy, and w is the energy endpoint of the inner bremsstrahlung. Then if we assume that this endpoint corresponds to a decay to the ground state of the resultant nucleus, the total decay energy will be given by the energy endpoint of the inner bremsstrahlung plus the binding energy of the K electron. The total number of bremsstrahlung photons emitted per disintegration is given by

$$\frac{N(\text{photon})}{N(\text{capture})} = \int_0^w \frac{N(k)dk}{N(\text{capture})} = \frac{a}{12\pi}(w/mc^2)^2$$

where a is the fine structure constant and mc^2 is the energy of an electron at rest. Then by a suitable normalization, we can say that the bremsstrahlung spectral shape is of the form $x(1-x)^2$, where x is a quantity proportional to k. If a portion of the bremsstrahlung spectrum is found experimentally,

then the total number of bremsstrahlung photons per disintegration can be found using this theory. For example, suppose the total number of counts under the curve is known from x equals 1 to x equals 0.65. We see that

$$\int_0^1 \frac{x(1-x)^2 \, dx}{x(1-x)^2 \, dx} \qquad \text{can be evaluated}$$

$$\int_{.65}^1$$

to yield 7.9. Then, by multiplying by this factor the total number of bremsstrahlung photons per disintegration is obtained. Various corrections can be made to this simple theory (Martin and Glauber, 1956); however, it is reasonable to assume that the simple theoretical results should be of the correct order of magnitude.

Chapter 2

APPARATUS

Sum Coincidence Spectrometer

For coincidence work a set of NaI(Tl) spectrometers was used. Most of this equipment has been described previously by Brown (Brown, 1964) and Ungrin (Ungrin, 1965) and this will not be repeated. Detectors used were Harshaw (NaI(Tl)) integral line detectors. One detector consists of a 1 3/4" by 2" crystal optically coupled to a 2 inch R.C.A. photomultiplier tube type 8053. The other has a 3" by 3" crystal directly coupled to a 3" photomultiplier tube type 8054. This use of different sized crystals is convenient in sum coincidence spectrometry (Brown and Roulston, 1965). The apparatus includes twin double - delay line amplifiers after a design by Chase and Svelto (Chase and Svelto, 1961). A cross - over pick - off coincidence system (ORTEC model 205) was used for the fast coincidence channel. The resolving time varied with window width of the single channel analysers from a low of about 20 nanoseconds for narrow windows to about 150 nanoseconds for both windows wide open. One of the problems encountered with this system was that careful setting of the cross - over pick - offs was required. Even so, due to a slight mismatching of the delay lines in the

amplifiers, there was a " walk " with pulse height variation of about 15 nanoseconds over the range of one amplifier, with the other amplifier held fixed. This was found using both an oscilloscope and a time-to-amplitude converter (NE 9110). A resolving time much longer than this was used so as to avoid trouble from the " walk ". This method had the advantage over a trigger circuit firing on all pulses in that energy discrimination was built into the fast timing circuit. With this circuit we were able to go to lower energies, the previous trigger circuit used by Brown and Ungrin not operating below 50 keV.

The sum coincidence experiments were performed using a two parameter Nuclear Data 4096 channel analyser (see fig. 1 a for block diagram). Our system is basically the same as that of Hoogenboom (Hoogenboom, 1959), with the exception that a fast timing circuit has been added. Instead of a single channel analyser determining the sum energy of the coincident gamma rays, a range of sums is displayed on the y-axis of the two parameter analyser. This is an important advantage in dealing with short lived sources.

Figure 1

Apparatus

a) Block Diagram of Sum Coincidence Spectrometer

TWO PARAMETER SUM COINCIDENCE

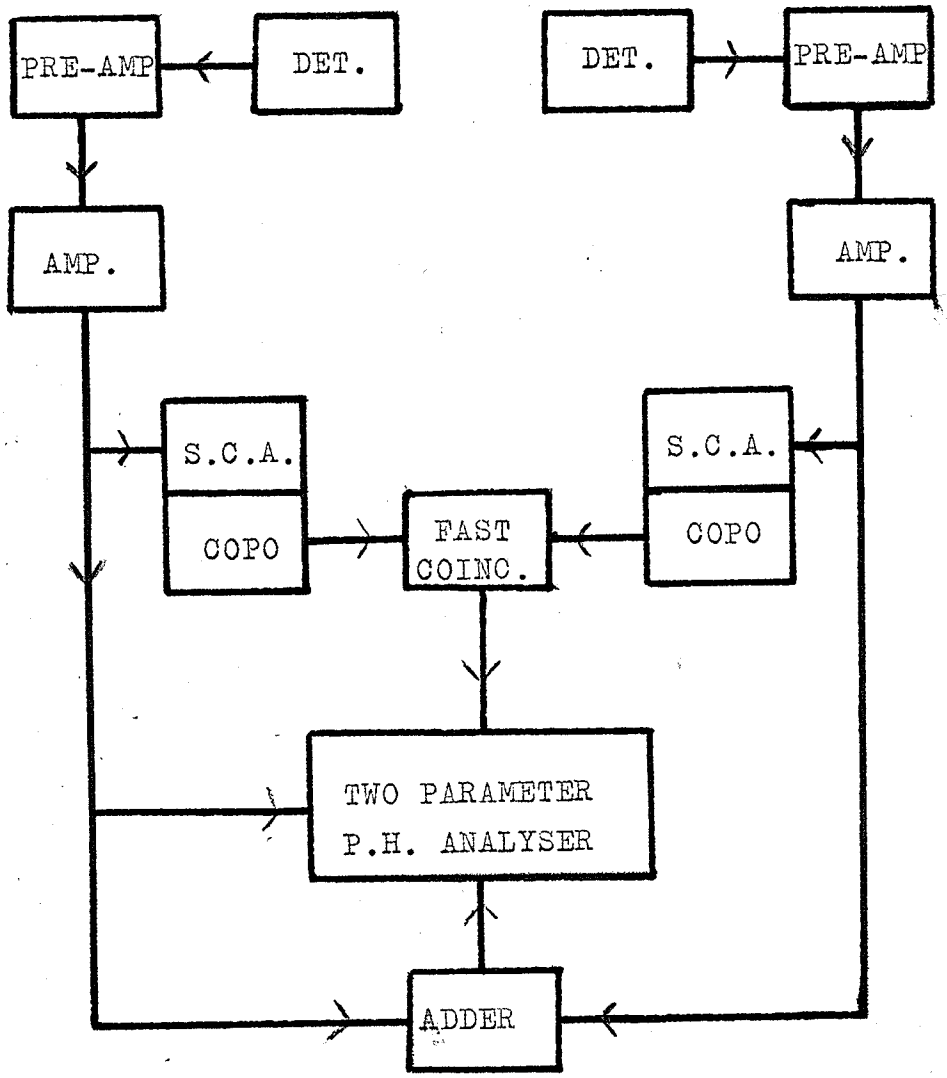
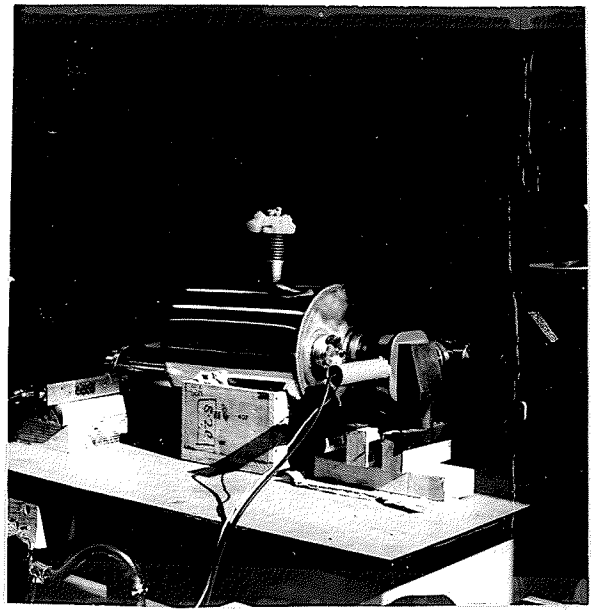


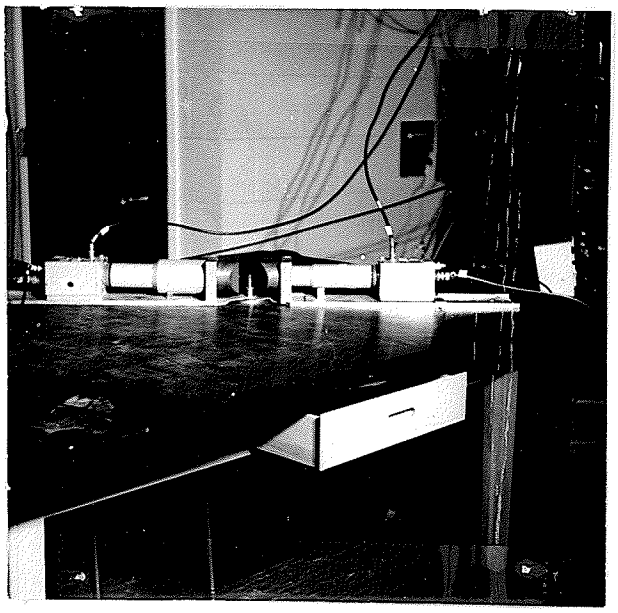
Figure 1

Apparatus

b) Cryostat and Detectors



Cryostat



NaI(Tl) Detectors

Lithium Drifted Germanium Detector

The normal resolution of a NaI(Tl) detector is 8%, although resolutions of 6% on the 661 keV Cs^{137} peak have been reported under good conditions. The principal limitation on the resolution attainable by scintillation detectors is the statistical variation in the number of photoelectrons emitted at the photocathode of the photomultiplier. This is due to the fact that it takes about 30 eV to produce an ion pair in NaI, in addition to the relatively low efficiency for conversion of light to electrons at the photocathode of the phototube.

One of the Chief advantages of using a solid state diode is that it requires only about 3 eV per ion pair in semiconductors such as germanium or silicon. This causes them to possess an inherently better resolution. The problem in the past has been the manufacture of diodes with sufficiently large depletion depths. The technique of making lithium drifted silicon diodes of sufficient depletion depth was mastered first. These were quite useful for stopping particles such as electrons, alphas, and protons. Because silicon has a low atomic number, these diodes were not very good for gamma ray spectroscopy, as the large Compton distribution obscured the small photopeaks.

Recently, the process of drifting lithium into germanium to form diodes has been mastered. Due to the higher atomic number of germanium, the cross section for the photoelectric effect is much higher. This enables them to be used at energies above 100 keV, which is the practical limit for photopeak production in silicon.

Because of the higher mobility of lithium in germanium as compared to silicon, it is necessary to operate these detectors below -40 °C. in order to keep the lithium from drifting out of the depletion region. For convenience, the usual practice is to operate them at liquid nitrogen temperature.

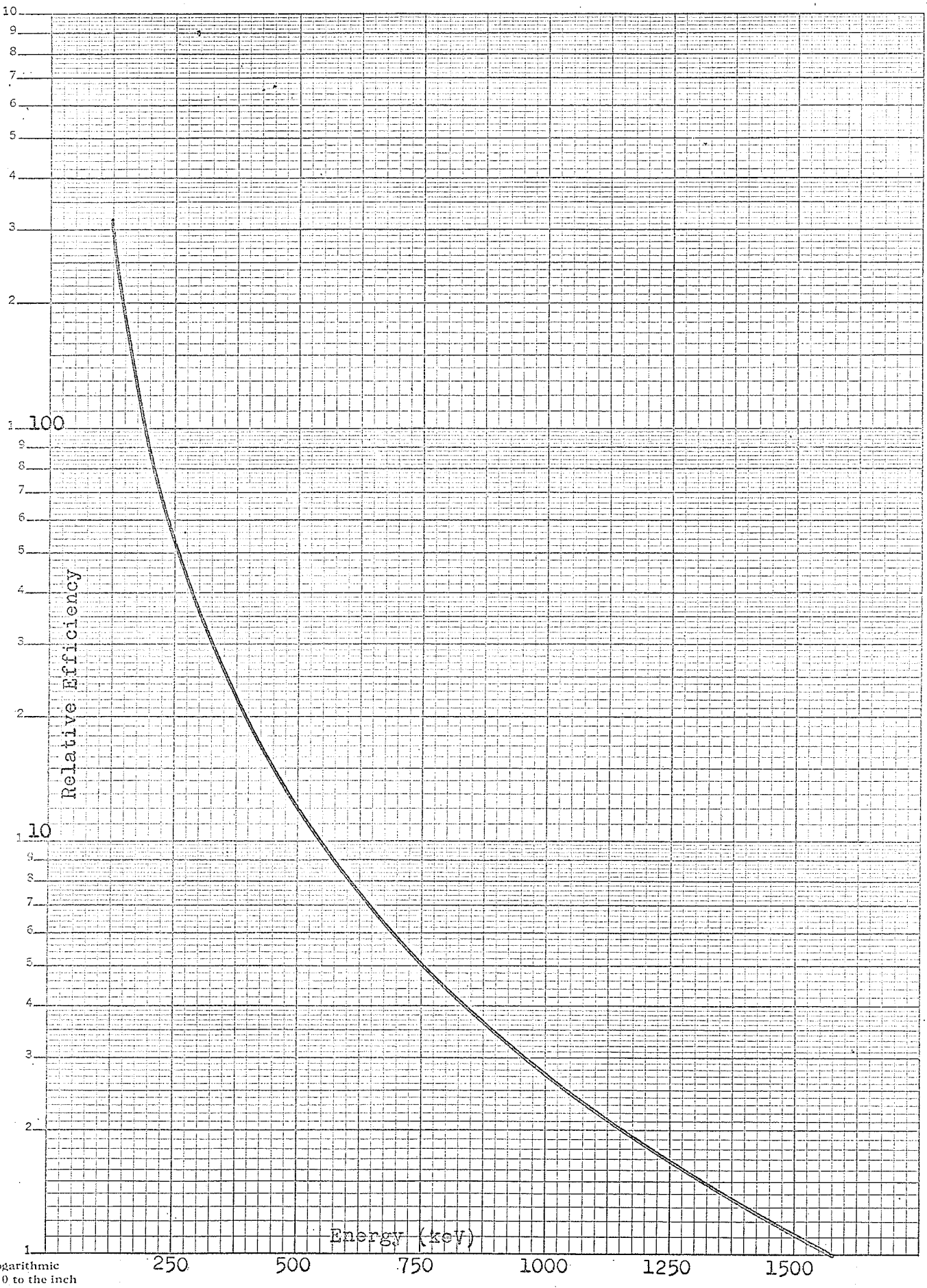
The detector used had an area of 2.8 cm^2 and a depletion depth of 2 mm. (R. C. A. type SJGG-2). It was kept in a cryostat containing liquid nitrogen. The cryostat used was kept evacuated using a model 921-0011 Varion " Vac-ion " pump which kept the pressure below 10^{-8} mm. of mercury. This prevented the detector leads from frosting up and slowed heat loss from the liquid nitrogen container. The preamplifier used was type NE 5231, and had a noise level of 2.1 keV at zero input capacitance and a noise slope of 0.04 keV/pF..

The spectrum was fed into a Nuclear Data 4096 channel analyser. The best resolution (full width of peak at half maximum height) obtained on the 122 keV peak of Co^{57} was 3.9 keV. After several months the detector inadvertently warmed up, due to vacuum failure. After it was repaired, it was found that the resolution had deteriorated to 4.3 keV on the 122 keV peak of Co^{57}. The leakage current of this detector was of the order of 10^{-9} amperes.

The efficiency of detection of gamma rays in the energy range below 1 MeV was quite low (of the order of 0.1 to 1%). The relative efficiency calibration of this detector was obtained using peaks of known intensity of La^{140}. A typical efficiency curve for one of the geometries used is included. As can be noted, the efficiency curve rises very sharply in the energy region below 200 keV, making efficiency calibration difficult in this region. It should be mentioned that the efficiency calibration was found to be dependent on the geometry used, particularly in the low energy region.

Figure 2

Ge(Li) Detector Efficiency Calibration



Semi-Logarithmic
Cycles x 10 to the inch

Collimation

One of the problems observed in analysing gamma spectra with the Ge(Li) detector was the intense positron annihilation radiation associated with the cyclotron-produced sources. This was particularly evident in the case of Zn^{62} with its many low energy gamma rays, which were to a large extent obscured by the Compton distribution due to the 511 keV photons. In an effort to reduce this positron radiation, several methods were tried. The first of these consisted of a 3" by 3" NaI(Tl) detector hooked to a single channel analyser whose output was used as an anti-coincidence gating pulse at the Nuclear Data analyser. The NaI(Tl) detector was mounted directly behind the source which was enclosed in a pyrex test tube, ensuring that no positrons would leave the region of the source. Since in positron annihilation the two photons produced are emitted in opposite directions, it was hoped that by firing the single channel analyser on all radiation detected by the NaI(Tl) crystal and putting it in anti-coincidence with Ge(Li) detector, the number of 511 keV photons recorded by the analyser from the Ge(Li) detector would be reduced. This worked to some extent, yielding a factor of improvement of perhaps four. One of the problems associated with this was that the

NaI(Tl) detector was not 100% efficient. A more difficult problem was that due to the low efficiency of the Ge(Li) detector (0.1 to 1%), high counting rates were necessary in the NaI(Tl) detector in order to obtain a reasonable counting rate in the Ge(Li) spectrum. This caused losses in the single channel analyser creating the anti-coincidence pulse.

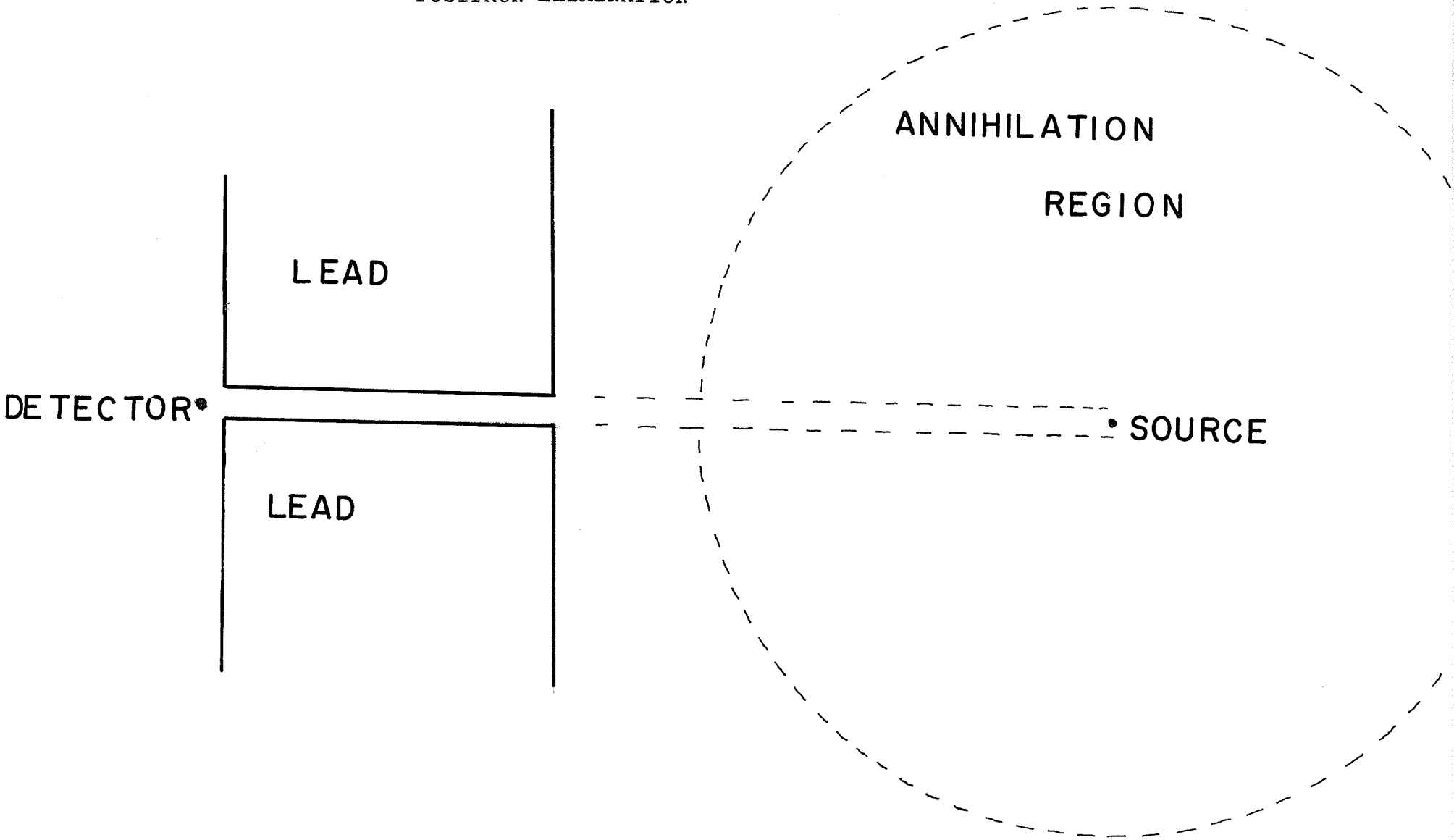
A more effective method developed consisted of the following. A lead collimator was used to expose only a portion of the total solid angle in space to the detector. The source used was a copper foil about 1 thousandth of an inch thick. This allowed most of the positrons to leave the source before annihilating (see Fig. 3). Since the Ge(Li) detector was only exposed to the actual copper foil, very few of the 511 keV quanta were able to strike the detector when the positrons annihilated. Experimentally it was found that a large number of positrons travelled at least six feet in air before annihilating. For this reason a vacuum chamber was not employed. Instead of shielding the detector with lead from all sides, it was found simpler to use a large glass box, which stopped the positrons before they could travel behind the lead collimating block and thus shower the detector from behind. This also had the advantage of sharply lowering the radiation

Figure 3

Block Diagram of Positron Annihilation Elimination System

level in the room. Since the sources necessary for this method to work well were of the order of 5 to 20 millicurie, this was not unimportant. This method yielded a reduction of annihilation radiation at the detector by a factor of about 40. From using sources of different thickness it is concluded that the limiting factor in this case was the annihilation of some of the positrons in the source itself. This was not, therefore, a fundamental limitation since thinner sources could have been made. However, the annihilation peak was now reduced to the same order of size as the peaks of the nuclear gamma rays, and for this reason, efforts to improve it further were not made.

POSITRON ELIMINATION



Chapter 3

RESULTS ON TIN - 113

There has been a good deal of interest in the past in the decay of Sn^{113} and some ambiguities exist, particularly in the energy spectrum above 400 keV. Philips and Hopkins (Philips and Hopkins, 1960) claim the existence of a 650 keV gamma ray with an intensity of about 6% of the intensity of the 255 keV transition. They got this result by means of a fast coincidence system, with the 24 keV K X-ray of indium in the other detector. Others failed to find such a peak and set upper limits to its intensity below that obtained by Philips and Hopkins.

This experiment was repeated, using the scintillation spectrometer previously described. The timing was by means of cross - over pick - offs firing on the cross - over of the double - delay line shaped pulses from the amplifiers. The resolving time of the system was about 80 nanoseconds as plotted out by inserting a variable delay in one side. The apparatus was checked for " walk " of cross - over timing pulses with pulse height using both an oscilloscope and a time-to-amplitude converter. The " walk " was found to be about 15 nanoseconds over the energy range

used. This would have produced a negligible effect. The X-ray channel was set to accept pulses in the energy range of 15 to 40 keV.

The experiment was repeated with two different Sn^{113} sources obtained commercially, one from Oak Ridge, and the other from Nuclear Science and Engineering Corp. Sources used had strengths of less than 1 microcurie. The detectors were brought close (within 3 inches) to the source in order to improve the signal to accidentals ratio of the coincidence count. The detectors were not shielded from each other, since any back scatter peaks would fall in the low energy region below 250 keV, which was not of interest in this experiment. The spectrum of the first source (from Oak Ridge) was obtained using a 100 channel CDC analyser (see Fig. 4a). The resulting spectrum shows the 255keV peak with the chance 393 keV peak about a factor of 15 less intense. In addition there are two other peaks at 600 and 800 keV. These are above the background, which was normalized to the 393 peak. We later repeated this experiment with the other source using a Victoreen analyser (see Fig. 4b). In this case we found that the 600 and 800 keV peaks were less intense relative to the 255 keV peak. In addition to this there was a peak at about 320 keV which was not

present on the previous spectrum. It was also noted that there was some evidence, despite poor statistics, for some higher energy peaks in the first spectrum. From this we can set an upper limit on the intensity of a possible 650 keV transition of 1/130 of the intensity of the 255 keV peak.

We also attempted to establish the total decay energy. Previous workers (Ungrin, 1965), using NaI(Tl) detectors, had fit the theoretical $x(1-x)^2$ shape of the inner bremsstrahlung to a singles spectrum of Sn^{113}. From this a bremsstrahlung endpoint of about 900 keV was found. Others, from conversion electron work (Durosinmi-Etti, Brundrit and Sen, 1965), tended to have lower values for the transition energy to the 393 keV level. These values seemed to be around 200 to 400 keV, although this was subject to large errors. Our coincidence work did not give us a good endpoint for the bremsstrahlung since the peaks at 600 and 800 keV obscured any underlying continuum. The relative intensity of the bremsstrahlung, as claimed by Ungrin in the energy region above 600 keV, was greater than anything found in our coincidence spectrum. From this we suspected that the high energy counts found by previous workers were due to some impurity.

In an effort to indentify these impurities in

Figure 4

Sn^{113} Spectrum Coincident With K x-ray

(NaI(Tl) Detector)

a) Seven Month Old Source

All Energies in keV

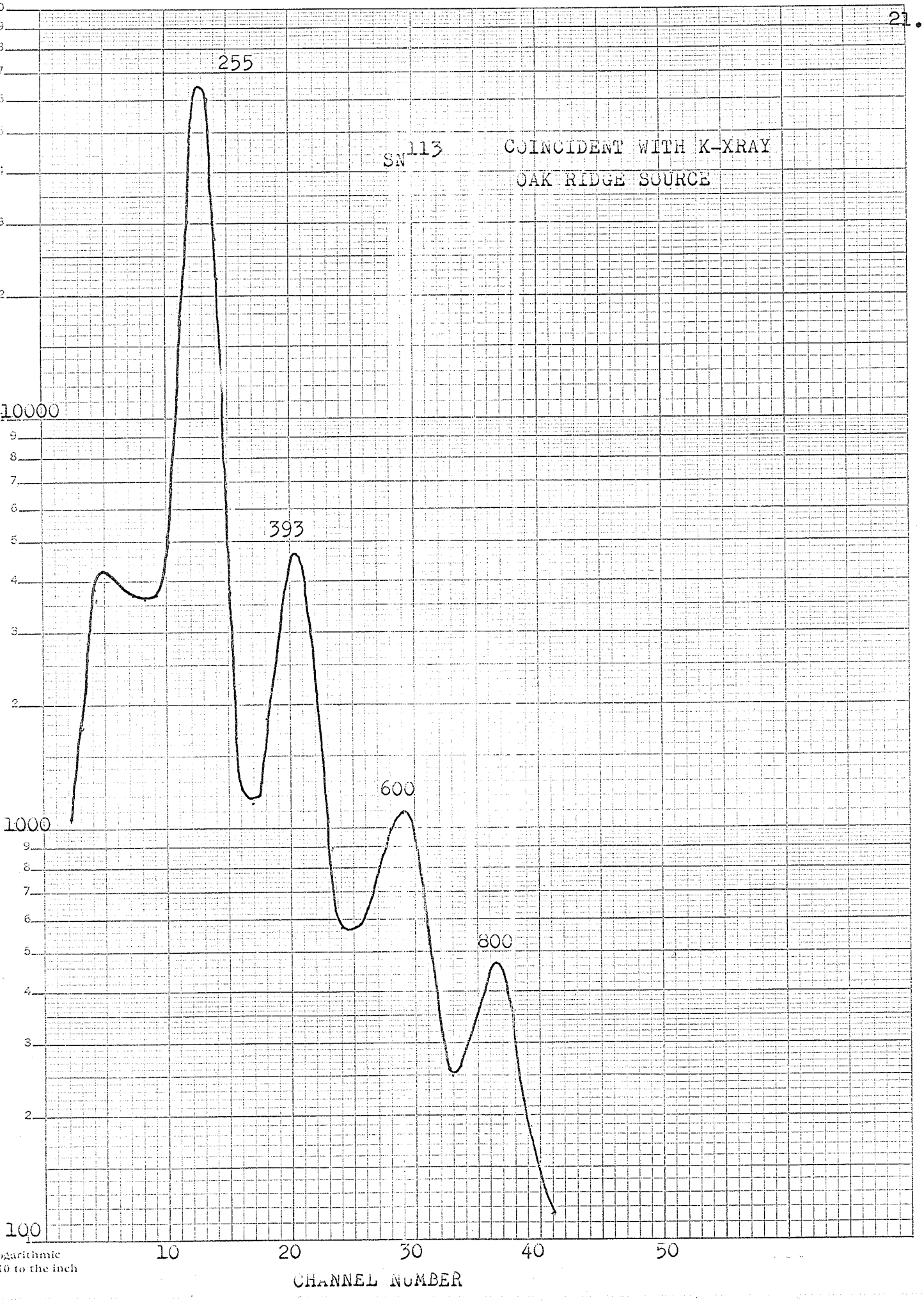

255
393
600
800
SN 113
COINCIDENT WITH K-XRAY
OAK RIDGE SOURCE
10000
1000
100
10
20
30
40
50
CHANNEL NUMBER
garithmic
0 to the inch

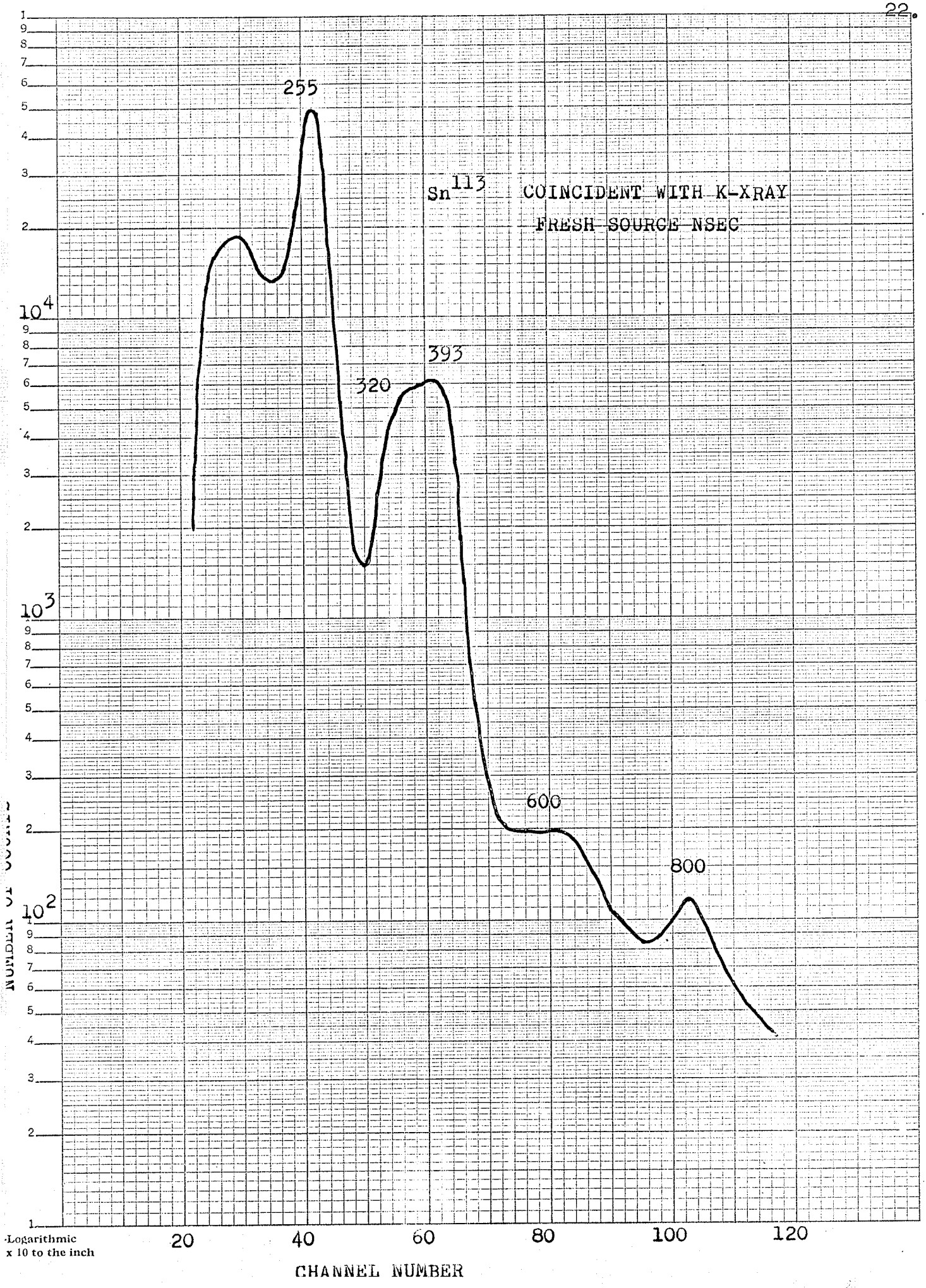
Figure 4

Sn^{113} Spectrum Coincident With K x-ray

(NaI(Tl) Detector)

b) Fresh Source

All Energies in keV


255
Sn^{113} COINCIDENT WITH K-XRAY
FRESH SOURCE NSEC
393
320
600
800
10^4
10^3
10^2
20
40
60
80
100
120
Logarithmic
x 10 to the inch
CHANNEL NUMBER

our sources and to pinpoint the bremsstrahlung endpoint, both sources were investigated using a 2.8 cm^2 by 2 mm depletion depth lithium drifted germanium detector.

The results of the Ge(Li) runs indicate that, although other impurities were present, Cs^{134} was rather prominent in both sources. This is indicated by a comparison of our spectra with those of Ewan and Tavendale (Ewan and Tavendale, 1965). This impurity was strongest in the Oak Ridge source, which is consistent with the fact that this source was about nine months old at the time the Ge(Li) spectrum was taken. If allowances are made for different rates of decay due to half-life, we find that both sources contain close to the same degree of Cs^{134} impurity (about 1 part in 9000). This leads us to conclude that Cs^{133}, which is the only stable isotope of cesium, must be commonly present as an impurity in the tin ore, the Cs^{134} thus being produced by a (n,γ) reaction.

This, in our opinion, explains Ungrin's results for the endpoint, since in a NaI(Tl) singles spectrum the upper tail of the 800 keV peak comes to about 950 keV. We know that the Cs^{134} was present in this source because it was the one before referred to as the Oak Ridge source. Also, due to the fact that Cs^{134} was found in both sources, we are inclined to believe that this explains

the results of the other measurements of the endpoint (Philips and Hopkins, 1960). This also explains the peaks as found in our coincidence spectrum, the 600 and 800 keV gamma rays being in coincidence with the external bremsstrahlung of the emitted beta rays and with the Compton distribution of other gamma rays. It was noted that there was no evidence for a 650 keV cross over transition. We were able to set an upper limit of 1/550 of the intensity of the 255 keV peak. This agrees with our coincidence data.

The Ge(Li) spectrum revealed a gamma ray continuum with an endpoint of 680 $\pm$ 10 keV as obtained from a Kurie plot (Fig. 5b). The intensity of this continuum was large enough to rule out the possibility of its belonging to Cs^{134}. While it is possible that it could belong to some other impurity, no gamma rays of sufficient intensity to suggest this were found. The fact that this bremsstrahlung was present in both sources tends to confirm that it belongs to Sn^{113}. The continuum was not a Compton distribution due to higher energy gamma rays since the peaks due to impurities were of much too low an intensity and since Sn^{113} possess no known gamma rays above 393 keV. There was enough of a Compton distribution present, particularly in the older source, to cause some error in the determination of the intensity of the bremsstrahlung.

A plot of the continuum corrected for detector efficiency is shown in figure 5a. From this we assume a probable endpoint to the continuum. Then if we plot $(\text{no. of counts}/x)^{1/2}$ versus $(1-x)$, we should get a straight line whose intercept will give a first order correction to our assumed value of the bremsstrahlung endpoint. In considering figure 5b it should be noted that the extreme points at each end have larger errors, due on one end to difficulties in background subtraction, and at the other to being so close to the 393 keV peak. Our final value for the bremsstrahlung endpoint is 680 ± 10 keV. This result was taken from the spectrum of the fresh source since here there was relatively less impurity present.

The only part of the bremsstrahlung considered was that above 393 keV. To get the total bremsstrahlung we assume a $x(1-x)^2$ shape and integrate over a portion of the bremsstrahlung spectrum. Our result for the fresh Sn^{113} source for the ratio of the intensity of the 255 keV gamma ray to the intensity of the bremsstrahlung was 1/3.3 with a possible error of 30%. For the older source there were more background difficulties due to the Compton distribution of the impurities. For this case our value for the ratio of the intensity of the 255 keV gamma ray to that of the bremsstrahlung was 1/1.8

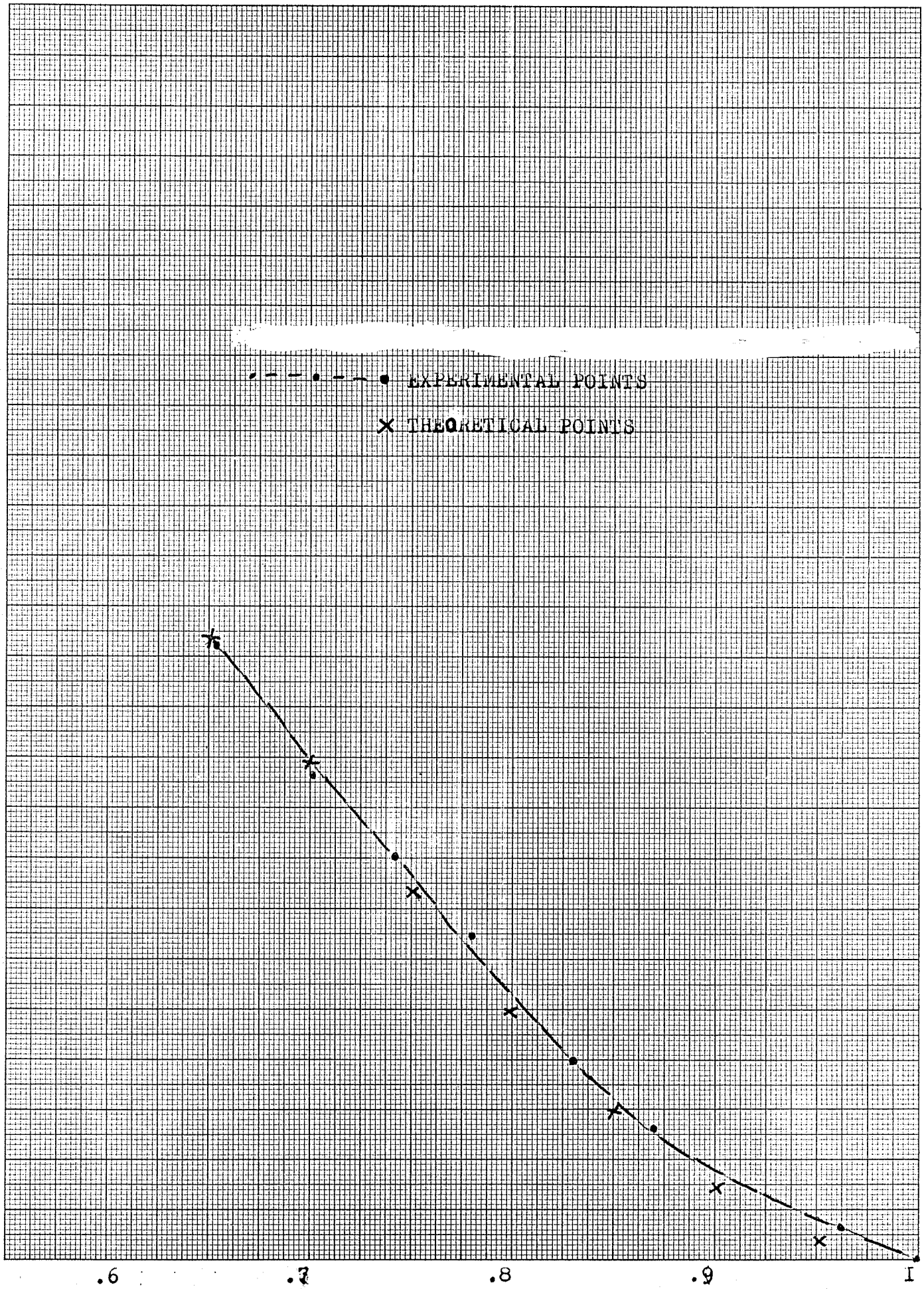
Figure 5

Bremsstrahlung Spectrum of Sn^{113} Using Ge(Li) Detector

a) Singles Display Corrected For Detector Efficiency

●- -●- -● Experimental Points

x x x x Theoretical Points

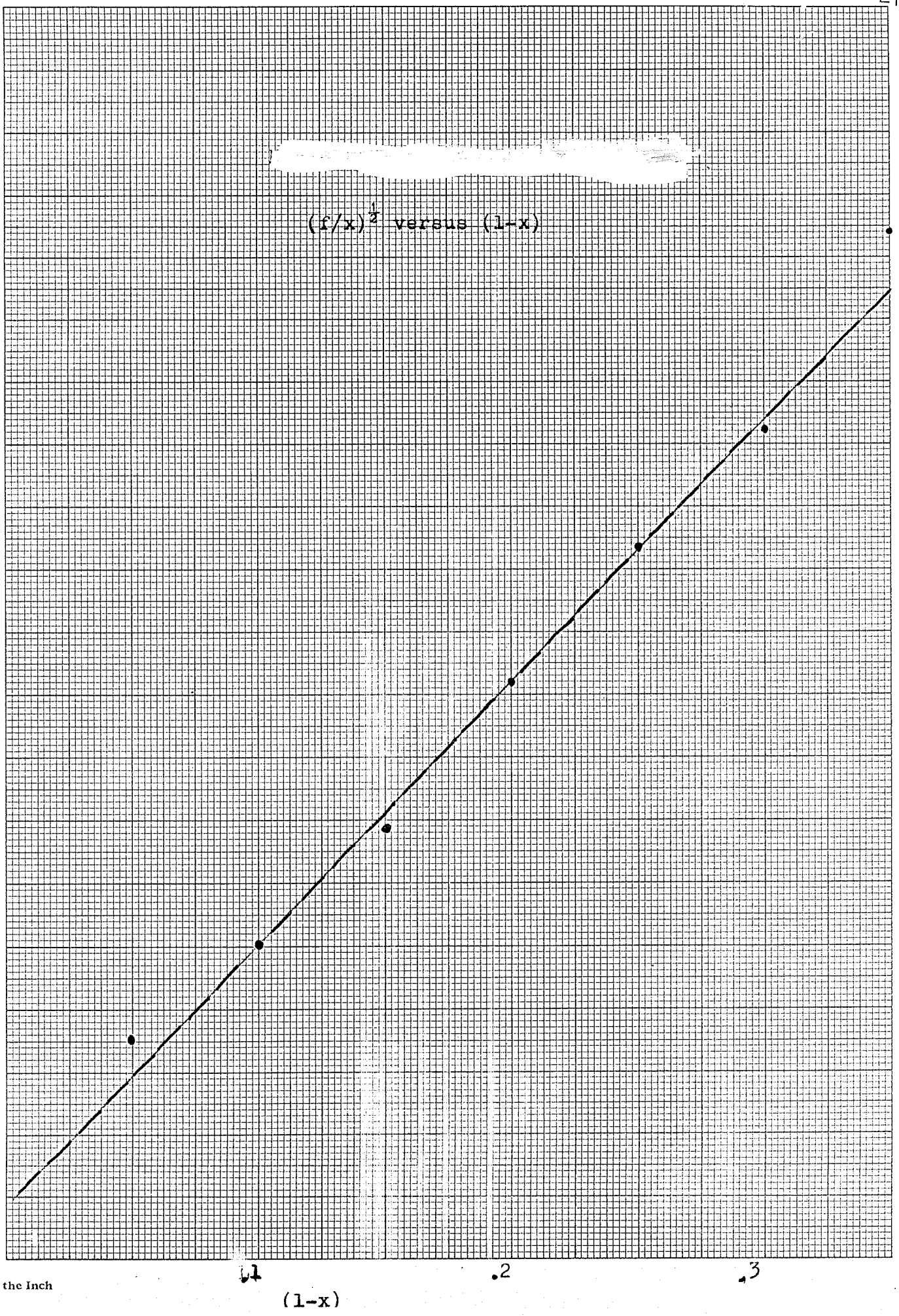


meters to the Centimeter

Figure 5

Bremsstrahlung Spectrum of Sn^{113} Using Ge(Li) Detector

b) Kurie Type Plot



ares to the Inch

with a possible error of 50%. The ratio of the intensity of the 255 keV gamma ray to that of the 393 keV gamma ray is 1/40 (Ungrin, 1965). If we assume 1/2 of the gamma rays are internally converted, then we arrive at the conclusion that there is emitted one bremsstrahlung photon for every 28 decays. Theoretically, for a bremsstrahlung endpoint of 680 keV, we have the result N(Brems.)/N(capture) = 1/3000. Our result differs from theory by a factor of 100. It might be noted that tin, with atomic number 50, possesses a closed proton shell, which might help to account for the discrepancy.

We were able to set an upper limit to the intensity of positron emission of 1/1000 that of the intensity of the 393 keV gamma ray. This is consistent with our estimate of the decay energy of Sn^{113} of 1.1 MeV (R. Bouchez and P. Depommier, 1960). Our value for the ratio of the intensity of the 255 keV gamma ray to that of the 393 keV gamma ray is 1/35 $\pm$ 15%. This is consistent with the work of others (Ungrin, 1965).

Chapter 4

CYCLOTRON PRODUCED ISOTOPES

Source Preparation

Electrolytic copper* of 99.98% purity was used as the target material for proton bombardment in the University of Manitoba cyclotron. The isotopic composition of the target was 30% Cu^{65} and 70% Cu^{63}. The irradiation procedure consisted of bringing one edge of the copper foil to the desired radius inside the cyclotron on a probe and then commencing the bombardment. Thickness of the foil as supplied was 0.005 inches. For some experiments thinner foils were necessary. In this case the foil was thinned by dipping it in nitric acid until the desired thickness was obtained. The energy of the bombarding protons was chosen from data given in ORNL-CPX-2 by McGowan, Milner and Kim. Beam currents used were in the microampere range.

The procedure employed in determining the source of the gamma radiation observed was as follows. Successive spectra were taken and the relative half-lives of the peaks determined. Because of the high resolution of the Ge(Li) detector it was relatively easy to see which isotopes were present, since it happened only rarely that two peaks were not resolved.

* Obtained from Fisher Scientific Company.

Table 1

Table of Isotopes Produced by Proton Bombardment of Copper

Proton Energy	Isotope Produced	Half-Life	Remarks
25 MeV	Cu^{62}	9.8 min.	One of main reaction products. Also decay product of Zn^{62}? $Cu^{63}(p,pn)Cu^{62}$
	Zn^{62}	9.3 hr.	One of main products. $Cu^{63}(p,2n)Zn^{62}$
	Zn^{63}	38 min.	Lower cross section, visible in spectrum for first few hours only. $Cu^{63}(p,n)Zn^{63}$
	Cu^{61}	3.3 hr.	Low cross section. $Cu^{63}(p,p2n)Cu^{61}$
	Cu^{64}	12.9 hr.	Lower cross section than main products. Only one gamma ray at 1.3 MeV. $Cu^{65}(p,pn)Cu^{64}$
	Zn^{65}	245 days	Only one gamma ray at 1.1 MeV. Only seen when other isotopes dead. $Cu^{65}(p,n)Zn^{65}$
40 MeV	Cu^{61}	3.3 hr.	One of main products.
	Cu^{62}	9.8 min.	One of main products. Also decay product of Zn^{62}
	Zn^{62}	9.3 hr.	One of main products.
	Zn^{63}	38 min.	Also produced.
	Cu^{64}	12.9 hr.	Also produced.
	Zn^{65}	245 days	Also produced.
	Zn^{61}	89 sec.	Probably produced. Not seen due to short half life.

This method proved adequate in the case of Zn^{62} where the most troublesome impurities were of much shorter half-life than Zn^{62}. Table 1 lists the isotopes observed. As stated in this table, there could have been short-lived isotopes present which were not observed, since it took about 15 minutes to get the source out of the cyclotron and to our detector.

In the case of Cu^{61} the situation was not quite so favourable as for Zn^{62}. Although all the previously established peaks were visible in the spectrum of the unpurified source, the large amount of impurity present, in particular Zn^{62}, was rather troublesome. Figure 9a is the best obtainable spectrum without a chemical separation being performed. This was obtained by using the Ge(Li) detector about one hour after the bombardment, thereby allowing time for the Cu^{62} to die away and not enough time for the longer lived Zn^{62} to show through too much.

To cut down on the amount of impurity present, an ion exchange column was set up using Dowex 1, 100-200 mesh, 4% cross-linkage, as the exchange resin to yield a categorical separation of zinc and copper. The bombarded copper foil was dissolved in concentrated nitric acid, giving cupric nitrate and zinc nitrate. The excess nitric acid was then boiled off, leaving a residue containing copper

and zinc nitrates. This was then dissolved in 9 normal HCl putting the Cu^{++} and Zn^{++} ions in solution. The solution was then poured into a 0.29 cm by 30 cm ion exchange column which had previously been washed with concentrated HCl. After the active solution was added, the column was washed with 0.5 normal HCl. The copper came out in the first 25 ml of wash, and the zinc came out after 60 ml. After this procedure, no sign of the Zn^{62} remained in the copper filtrate, until the Cu^{61} had been allowed to die off for over 48 hours.

Zinc - 62

Zn^{62} was obtained by means of a Cu^{63} $(p,2n)Zn^{62}$ reaction using 25 MeV protons on a natural copper target. Because Zn^{62} decays to Cu^{62} which is unstable, a pure Zn^{62} source is unattainable. Since Cu^{62} can also be produced directly by means of a Cu^{63} $(p,pn)Cu^{62}$ reaction, however, it was possible to sort out the two spectra. Due to the Cu^{65} which comprised 30% of the bombarded foil, Zn^{65} and Cu^{64} were also produced. Due to the long half-life of Zn^{65} (245 days), it was only observable long after the irradiation. Zn^{63} and Cu^{61} were the only other impurities observed.

The experimental procedure was to look for the more intense gamma rays of suspected impurities. If these were absent, it was assumed that these impurities were absent. In this case it was found that from about 3 hours after irradiation until about 40 hours there was very little impurity present. The only peaks of different half-life discovered during this time were due to Cu^{61} , and these were very weak. After 40 hours the only gamma ray of Cu^{64} at 1.3 MeV started to show through. Days later, the only gamma ray of Zn^{65} at 1.1 MeV became visible.

Because of the low efficiency of the Ge(Li) detector at higher energies, a singles NaI(Tl) spectrum

was run on a source 46 hours after the bombardment in order to check for long lived activities. The resulting spectrum (Fig. 6) shows the 1.3 MeV peak due to Cu^{64} about 1/15 the intensity of the 599 keV peak of Zn^{62} . Two peaks at 880 and 1170 keV are attributed to Cu^{62} which is in secular equilibrium with the Zn^{62} . Although Zn^{65} possesses a 1.1 MeV gamma ray, the activity due to it is too weak to contribute anything at this stage. Since any possible gamma rays which are not listed in the literature as belonging to Cu^{64} or Zn^{65} would necessarily be of low intensity, these impurities can be safely ignored. Because the Cu^{61} which is weakly present as an impurity has a spectrum which is well known to us, it too should cause no trouble. For this reason no chemical separations were undertaken in connection with the Zn^{62} work.

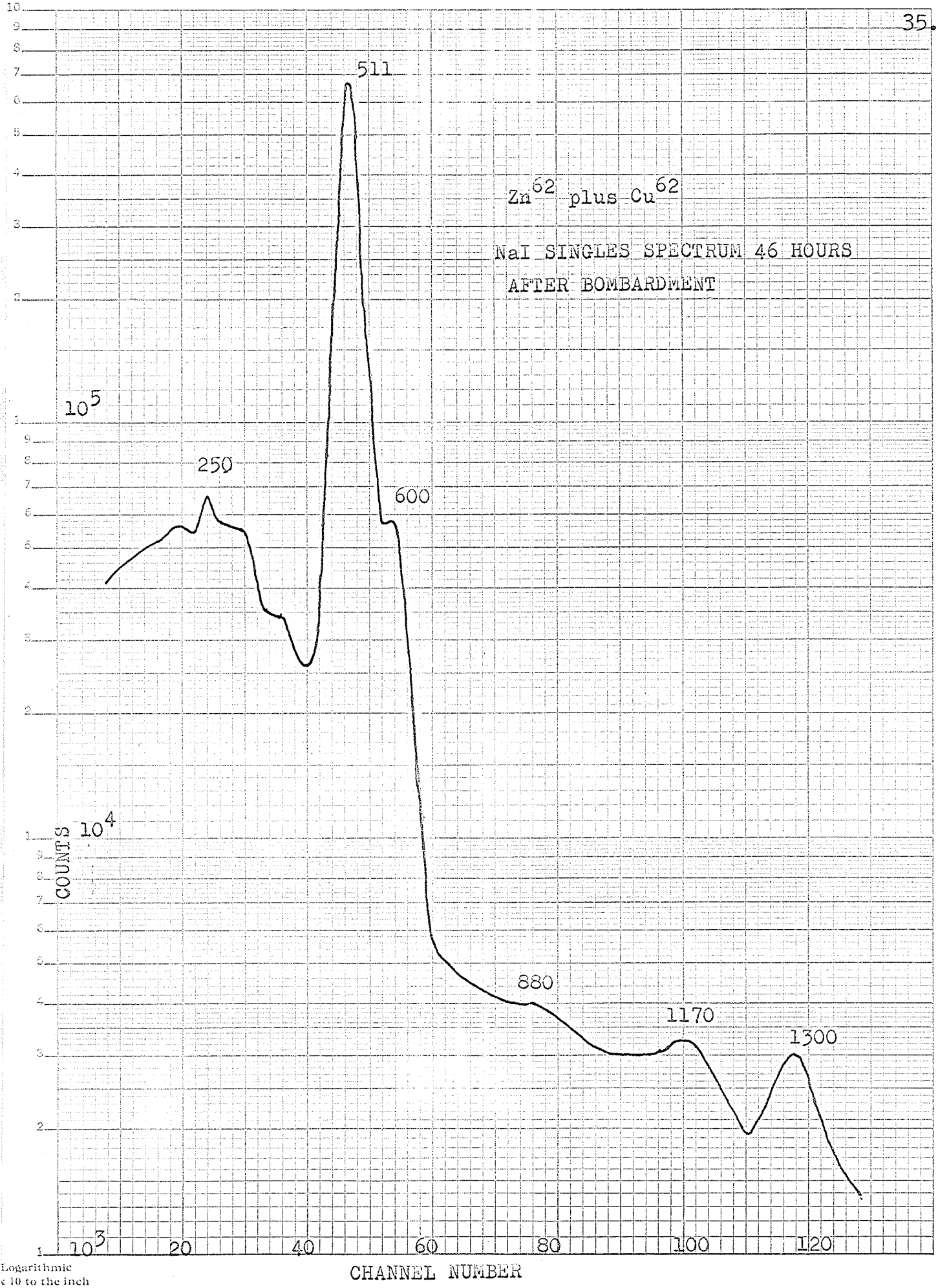
A set of $\gamma - \gamma$ coincidences has been done by Brun et al (Brun et al, 1957), and this was not duplicated. However, NaI(Tl) detectors were employed in the sum coincidence manner using a 2-parameter Nuclear Data analyser in an attempt to check out the decay schme as presented by them, and to trace out decay sequences.

The results are listed in the table below. The results are in agreement with the work of Brun. Equipment was set up as shown in figure 1a, and as

Figure 6

Singles Spectrum 46 Hours After Bombardment of Copper Foil With 25 MeV Protons (NaI(Tl) Detector)

All Energies in keV



Logarithmic
x 10 to the inch

described earlier. Resolving time of the fast coincidence circuit was about 150 nanoseconds. Different portions of spectra were looked at by changing either levels of single channel analysers, or by change of gain at the analyser.

Table 2

Summary of Coincidence Work

ENERGY (MeV)	IN COINCIDENCE WITH (MeV)		
0.25	0.25	0.53	0.40
0.53	0.040	0.25	
0.040	0.53		
0.40	0.25		

Error in energy determination was 0.05 MeV in all cases.

Several of the energies listed here could belong to one or several gamma rays as found with the Ge(Li) detector. In particular the 0.53 MeV energy listed could be either nuclear or annihilation radiation. Superimposed upon this was the Compton distribution due to the intense annihilation radiation. The detectors were placed at right angles to each other, so that the source was not between them. However, due to this source of chance coincidences and spurious coincidences due to multiple scattering of 2 annihilation quanta in

combination with the relatively low resolution of NaI(Tl) detectors, we did not see any of the peaks discovered by means of the Ge(Li) detector in this way.

Numerous Ge(Li) singles spectra were run on Zn^{62} plus Cu^{62} . The half-life of peaks found was measured relative to the 599 keV peak by taking spectra of the source at various times after the irradiation. In addition to this, the absolute half-life of the 599 keV peak was measured. Due to the low efficiency of the detector and to the large number of counts coming in at other energies, we were unable to get a very high counting rate in this peak. Because of this, the method used was to run the entire spectrum for ten hours, read out the data, and then continue counting for another ten hours. In this way the radiation was followed through four successive half-lives. The counting rates used were fairly high initially, and because of this, the dead time of the analyser had to be corrected for. This was done by using a clock which fired a pulse into the analogue to digital converter of the analyser approximately every 70 milliseconds to record the total " live time " on the analyser. Then, if both " live time " and actual time elapsed are known for each of the ten hour runs, the dead time of the analyser can be corrected

Figure 7

Singles Spectrum of Zn^{62} Plus Cu^{62}

(Ge(Li) Detector)

a) Without Positron Elimination

All Energies in keV

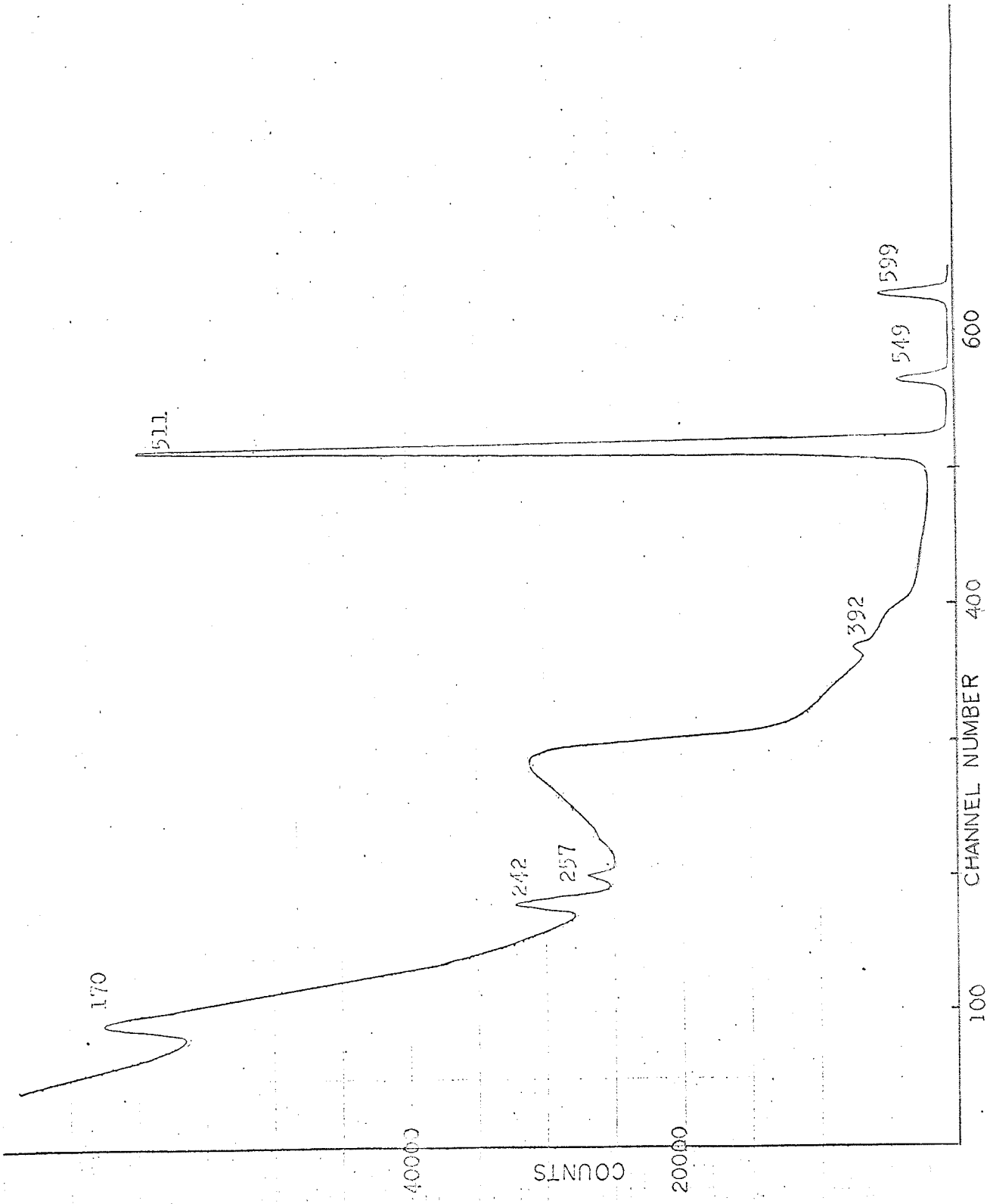

170
242
257
392
511
549
599
COUNTS
40000
20000
100
400
600
CHANNEL NUMBER

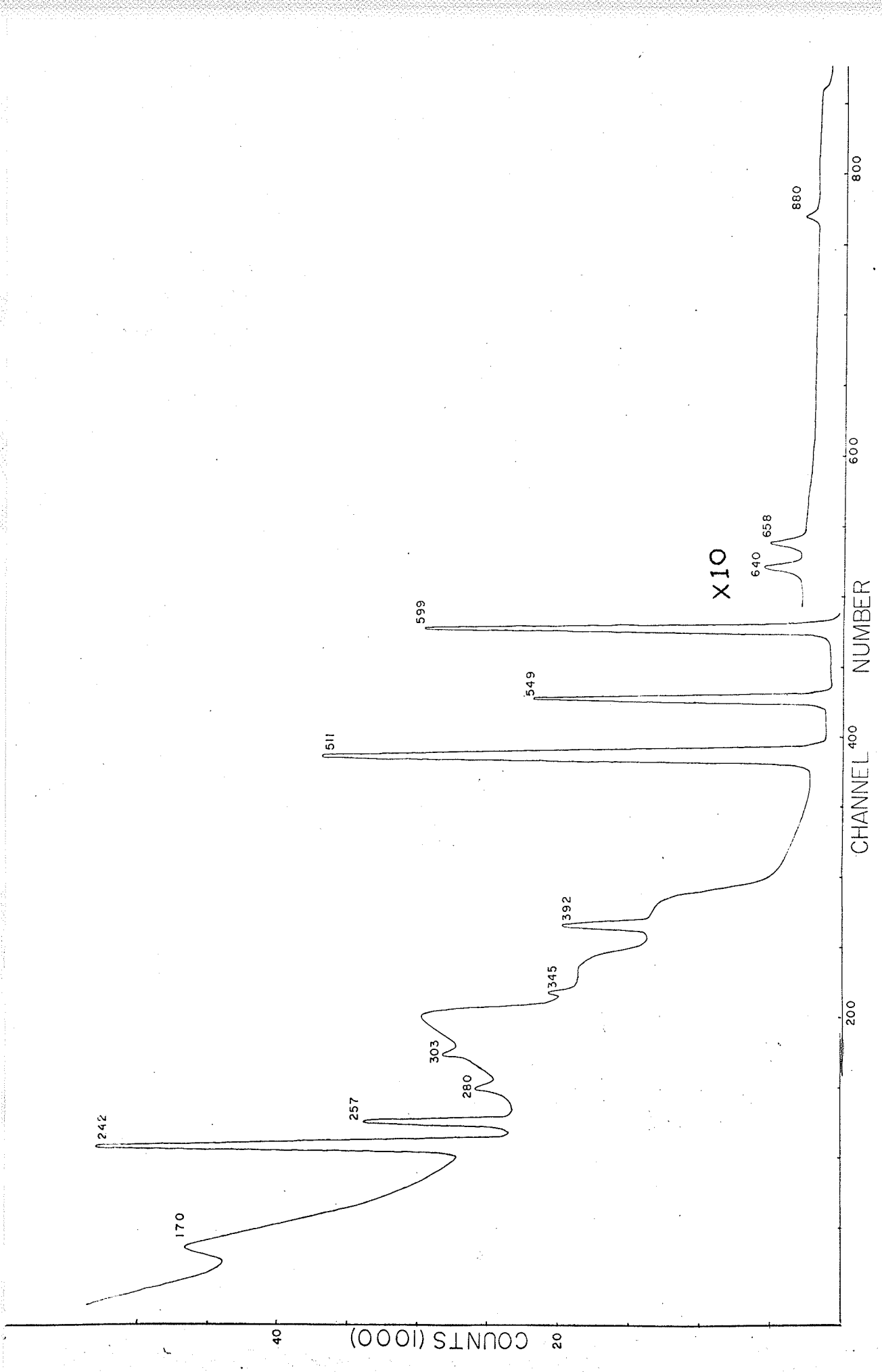
Figure 7

Singles Spectrum Of Zn^{62} Plus Cu^{62}

(Ge(Li) Detector)

b) With Positron Elimination

All Energies in keV


COUNTS (1000)
40
20
CHANNEL NUMBER
200
400
600
800
170
242
257
280
303
345
392
511
549
599
640
658
880
X10

for by multiplying the area of the chosen peak by:

$$\frac{\text{Actual time}}{\text{Live time}} .$$

The counting rates used were about 10,000/sec., well below the limit of the preamplifier and amplifier, so no further correction was applied for these. Then, if B equals the ratio of the two areas under the peaks, for two successive ten hour trials we get the equation:

$$\exp(-20x) - \frac{1}{B} \exp(-10x) = \frac{B-1}{B}$$

where x is the decay constant. Then the half-life can be determined. From this we got the result that the half-life associated with the 599 keV peak was 9.3 ± 0.2 hours. This is in good agreement with the work of Nussbaum et al (Nussbaum et al, 1954), whose value for the half-life of Zn^{62} was 9.33 hours.

The best spectrum of Zn^{62} plus Cu^{62} was obtained using the collimation method of eliminating the annihilation radiation. The relative half-lives of the weak transitions were determined as well as statistics would allow from successive collimated runs. In addition, the 345 keV gamma ray was first resolved in a singles NaI(Tl) spectrum using an anti-Compton cone. This enabled us to establish its half-life more accurately.

Table 3 gives a list of energies, intensities, and estimates of errors in measurement of half-life relative to the 599 keV peak. As noted in table 3, there is evidence for a widening of the peak at 511 keV. This was taken as due to the 510 keV nuclear gamma ray as found by Brun et al in his coincidence work. In addition to peaks listed as having a 9.3 hour half-life, there are two shorter lived peaks to be seen. These could be identified from their energies and relative intensities as belonging to Cu^{61}. No evidence for other peaks of noticably different half-life was found. The 880 and 1170 keV peaks were attributed to Cu^{62}. This follows from our work and from previous investigations done on Cu^{62}. It is, of course, possible that any of the weak peaks found in the singles spectra with the Ge(Li) detector could belong to Cu^{62}. However, from elastic scattering work and from previous investigations of Cu^{62} this appears doubtful. At any rate, we could not find them in later work on Cu^{62}. Using our data and that of others, we then could set up a decay scheme of Zn^{62}. This decay scheme is different from that of Brun in several important respects. In particular, our estimate of the intensities of the 242 and 257 keV peaks is radically different from his estimate, giving a reversal of the order of emission. The upper limit for the

Table 3

Table of Zn^{62} Plus Cu^{62} Gamma Rays

Energy(keV)	Half-Life*	Intensity	Remarks
640 ± 2	$6 < T_{\frac{1}{2}} < 11$ hr.	0.4 ± 0.06	
599 ± 1	T_{599}	27 ± 3	Absolute half-life of this peak determined to be 9.3 0.2 hr.
549 ± 1	T_{599} ± 0.5%	18 ± 2	
510 ± 5	not resolved	14 ± 5	This intensity taken from Brun's data.
392 ± 1	T_{599} ± 0.5%	2.9 ± 0.3	
345 ± 3	T_{599} ± 1%	0.35 ± 0.05	Seen in NaI(Tl) singles spectrum and later with Ge(Li) detector.
303 ± 3	T_{599} ± 25%	0.24 ± 0.04	
257 ± 1	T_{599} ± 0.5%	2.4 ± 0.4	
242 ± 1	T_{599} ± 0.5%	7.0 ± 1	
42		41 ± 7	Not seen in Ge(Li) spectra due to low energy. This intensity taken from Brun's data.
880 ± 9		0.15 ± 0.03	
1170			Compton edge due to it seen. Peak went over end of analyser.

* Half-life of peaks measured relative to that of 599 keV peak

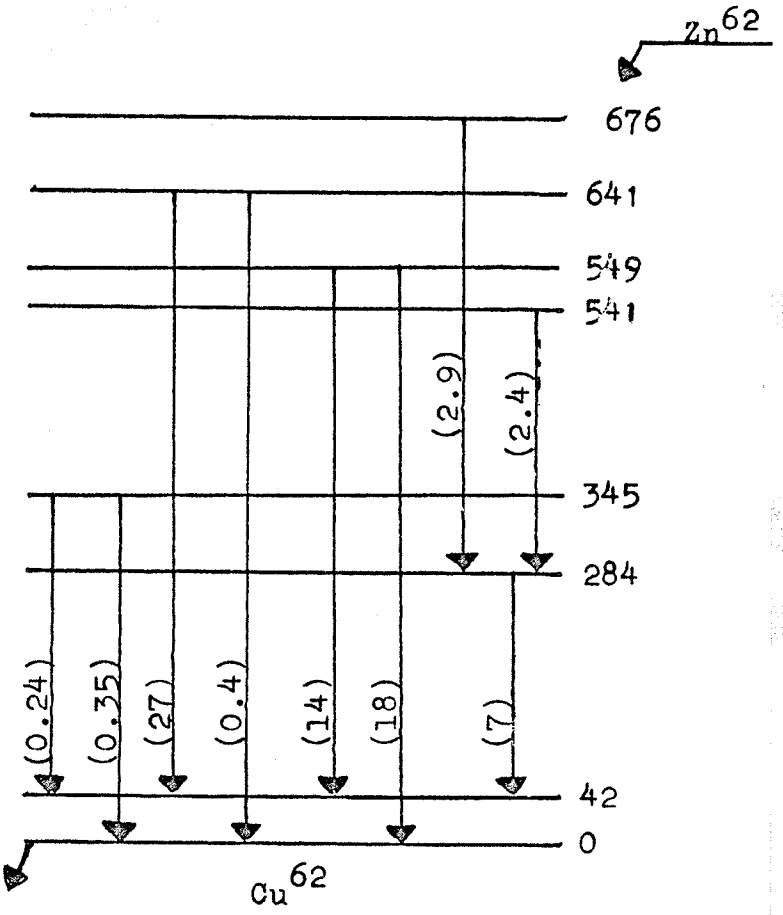
intensities of peaks with energy greater than 600 keV and less than 1000 keV and not listed in the literature is about 0.01%. Below 345 keV there exists a large Compton distribution which makes matters more difficult A rough upper limit for transitions between 80 and 345 keV which have not been mentioned would be 0.02%. In setting up a decay scheme we are faced with the problem that we are unable to find many of the peaks in the coincidence work due to the lack of resolution of the NaI(Tl) detectors. The low efficiency of the Ge(Li) detector made coincidence work impractical. A tentative decay scheme making use of the fact that many of the gamma rays can be arranged into pairs having difference in energy of 42 keV has been set up (see Fig. 8).

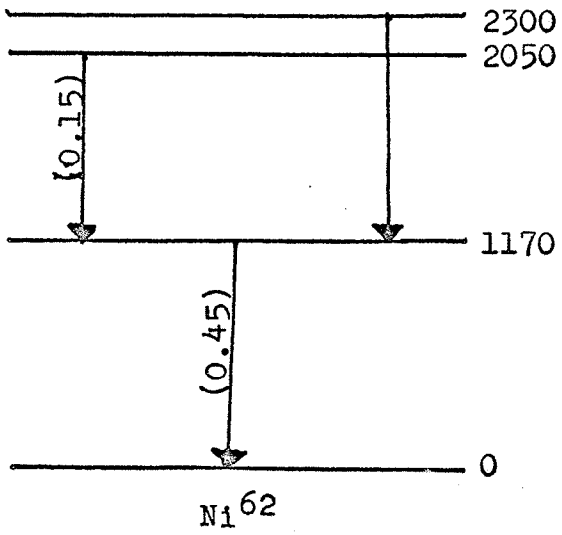
Figure 8

Decay Scheme Of Zn^{62} And Cu^{62}

All Energies in keV

Intensities given in brackets


Zn62
676
641
549
541
(2.9)
(2.4)
345
284
(0.24)
(0.35)
(27)
(0.4)
(14)
(18)
(7)
42
0
Cu62


2300
2050
(0.15)
1170
(0.45)
0
Ni62

Copper - 62

This isotope possesses an intense positron radiation, branching about 97% to the ground state. For this reason the gamma rays associated with Cu^{62} are quite weak. The levels of stable Ni^{62} have been determined by elastic scattering experiments and from the decay of Co^{62}, as well as from the radiation of Cu^{62}. From this the levels of Ni^{62} are fairly well known, but the gamma rays attributed to the decay of Cu^{62} remain uncertain (Butler and Gossett, 1958 a).

Some of the data obtained was from Zn^{62} in secular equilibrium with Cu^{62}. From this, two gamma rays were found, one at 880 keV and the other at 1170 keV. The ratio of the intensity of the 599 keV gamma ray of Zn^{62} to that of the 880 keV gamma ray of Cu^{62} is 180:1.

The work on Cu^{62} produced directly by a Cu^{63} $(p,pn)Cu^{62}$ reaction was hampered by the large amounts of impurities present. Due to its short half-life (9.8 min.), chemical separation was attempted on this source. Since one of the impurities was Cu^{61}, this would have proved difficult in any case. The procedure employed was to run several successive spectra in order to find out which of the peaks were short lived. By this method peaks at 880 keV and at 1170 keV were seen. The relative intensity of the 880 keV transition to that of the

1170 keV peak was 1/3 with a possible error of 50%. No 1130 keV gamma ray attributed to Cu^{62} by Butler and Gossett (Butler and Gossett, 1958a) was seen. However, the statistics were very poor, and for this reason no upper limit on the intensity of the 1130 keV transition was set.

Copper - 61

This isotope was produced with 40 MeV protons, using the internal beam of the cyclotron, by means of a Cu^{63} (p,p2n)Cu^{61} reaction. This energy is well above the threshold for a (p,n) reaction, and as expected, no Zn^{63} was detected in this spectrum. However, Zn^{62} and Cu^{62} were present to a large extent. Figure 9a shows the best obtainable spectrum using the positron annihilation radiation reduction method for the source as obtained from the cyclotron.

The zinc isotopes were then eliminated, using the ion exchange column as previously described. A series of Ge(Li) spectra was then run, one after the other, in order to check the relative half-lives of the peaks. The decay of Cu^{61} was followed through five successive half-lives.

Previous work on the levels of Ni^{61} has been done by several methods. A summary of work on the levels of Ni^{61} has been published by Butler and Gossett (Butler and Gossett, 1958 b). In work on the purified copper sources, the collimation method could not be used very effectively. The reason for this is that it proved rather difficult to get sufficient activity through the ion exchange column. For this source, the relative amount of positron emission is not as large as in the

Figure 9

Singles Spectrum of Decay of Cu^{61}

(Ge(Li) Detector)

a) Before Chemical Separation Performed on Source

All Energies in keV

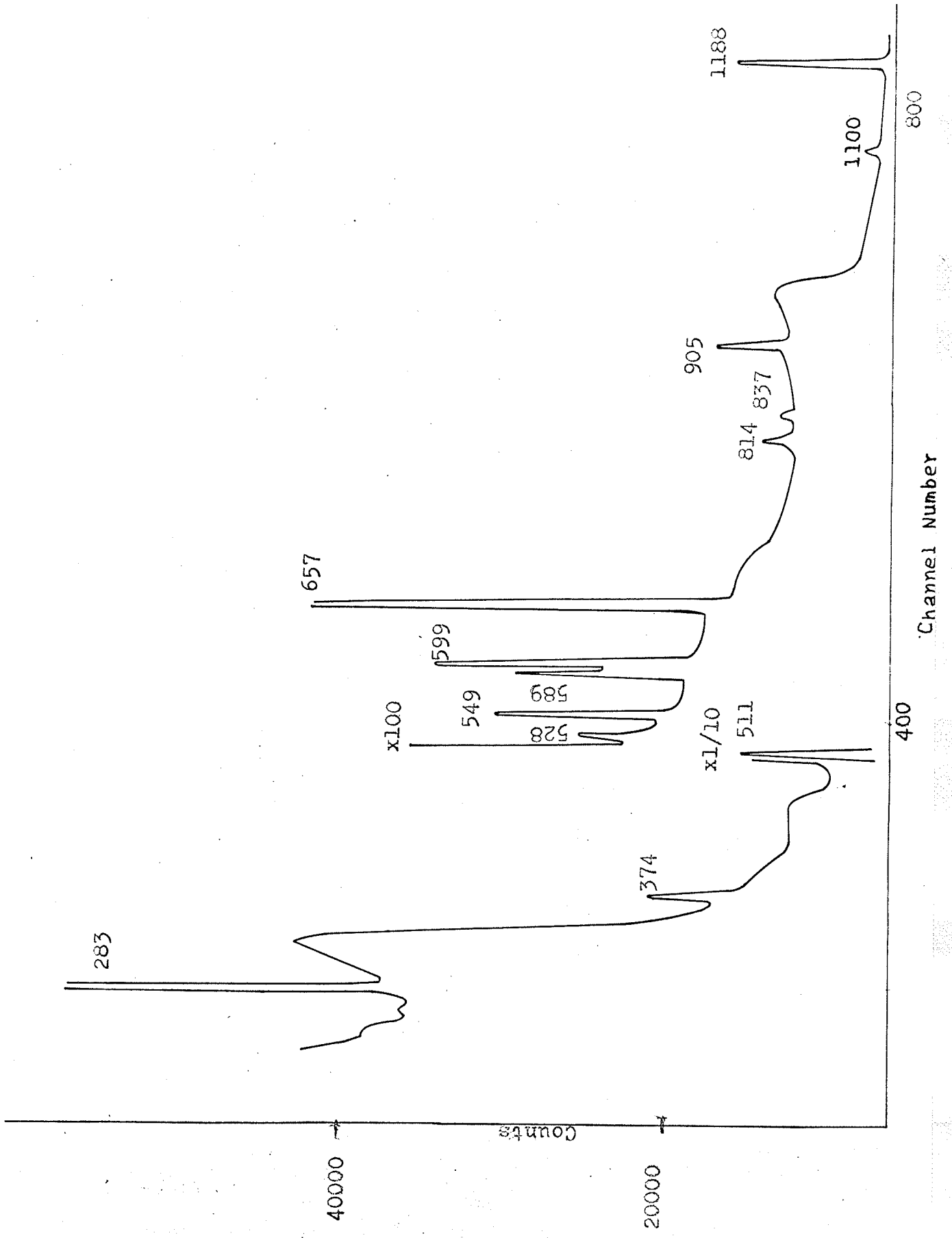

40000
20000
Counts
283
374
x1/10
511
x100
549
528
589
599
657
814
837
905
1100
1188
400
800
Channel Number

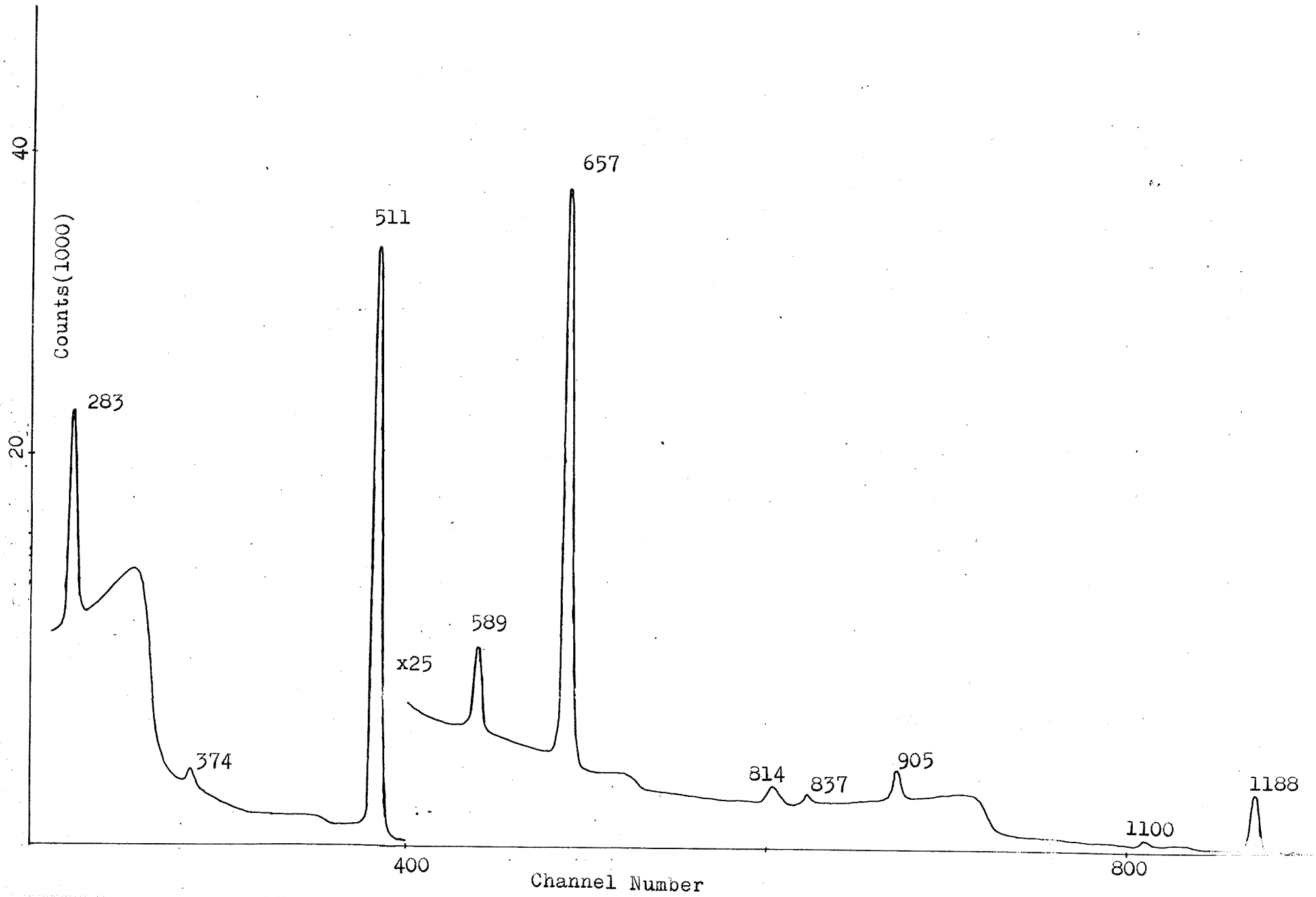
Figure 9

Singles Spectrum of Decay of Cu^{61}

(Ge(Li) Detector)

b) After Chemical Separation Performed on Source

All Energies in keV


40
20
Counts(1000)
283
374
511
589
x25
657
814
837
905
1100
1188
400
800
Channel Number

case of Zn^{62} plus Cu^{62} . Because of this, the positrons did not pose so much of a problem.

The impurities present even after the chemistry was performed include Cu^{64} , identified by its only known gamma ray at 1.3 MeV. This was quite weak, the amount of impurity present being of the order of 1/100 the intensity of the 657 keV peak of Cu^{61} . Another peak of longer half-life was found at 814 keV. The half-life of this gamma ray was estimated to be between 8 and 15 hours. Its intensity was of the order of 1/10 that of the 657 keV gamma ray of Cu^{61} . Two other peaks at 528 and 837 keV, not listed by Nussbaum et al (Nussbaum et al, 1956) were found. These had a half-life quite close to that of Cu^{61} . The accuracy of the estimate of their half-life is limited by the statistics in the number of counts, and the estimated error is about 50%. These peaks were followed through three Cu^{61} half-lives without any noticable change in their relative intensities. The rest of the spectrum agrees with that of Nussbaum et al (Nussbaum et al, 1956), although our energy and intensity measurements are more precise in most cases. The list of gamma rays and intensities is given in table 4, with an estimate in the table of possible error in relative half-life, although almost all the transitions have been previously established as belonging to Cu^{61} . A decay

scheme similar to that of previous experimenters is given. The 528 keV peak fits into the previously proposed decay scheme. More accurate energy determinations of the 1100 keV gamma ray make it doubtful that it fits in where it was previously thought to be. A new level has been tentatively added at 1100 keV to accommodate this gamma ray, although we have no coincidence measurements to locate its position in the decay scheme. The 837 keV gamma ray has not been included, since we did not have enough data to indicate its position.

Figure 10

Decay Scheme of Cu^{61}

All Energies in keV

Transitions In Ni^{61} Following Decay Of Cu^{61}

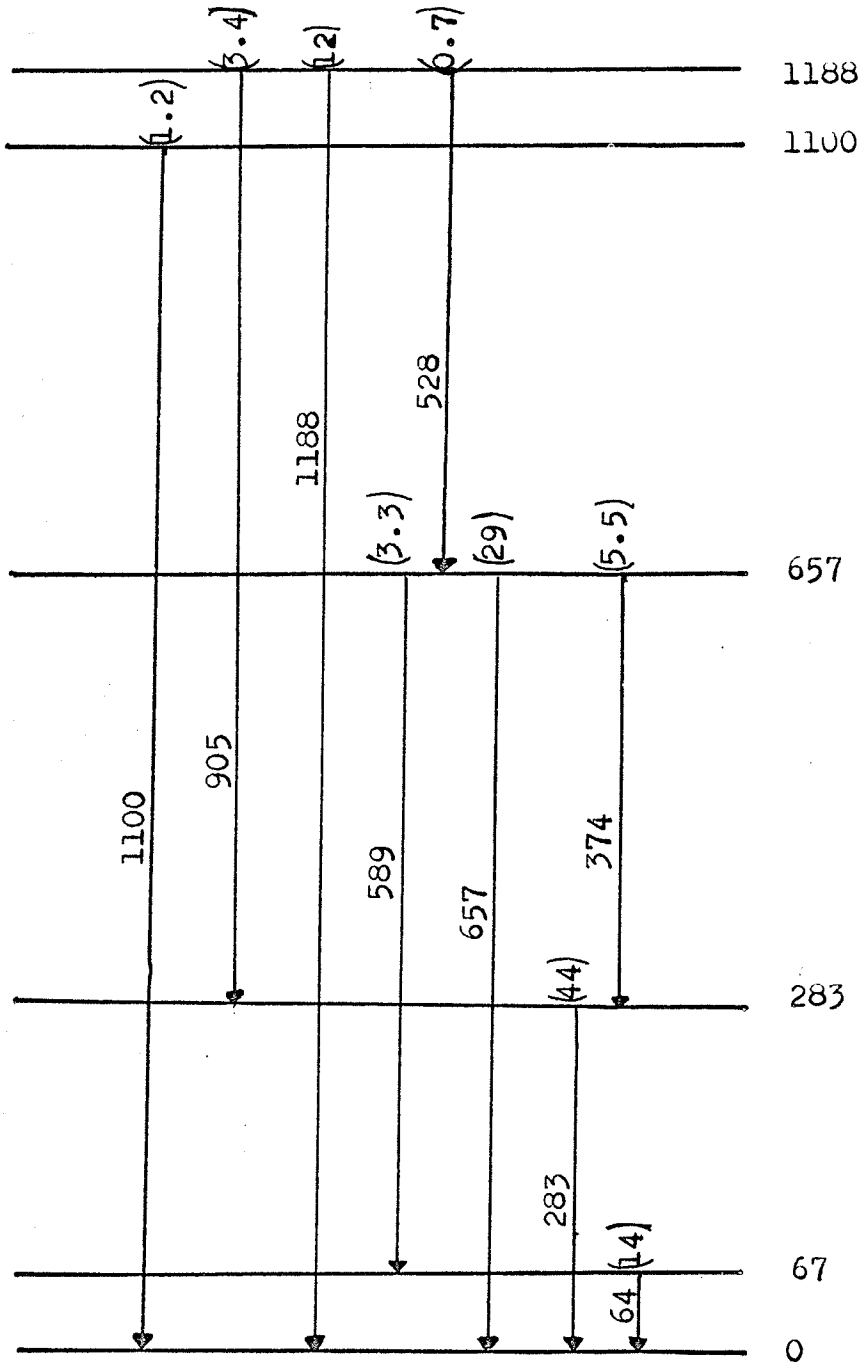


Table 4

Table of Cu^{61} Gamma Rays

Energy(keV)	Half Life(hr.)*	Intensity	Remarks
1188 ± 5	3.3 ± 1%	12 ± 2	
1100 ± 8	3.3 ± 10%	1.2 ± 0.2	
905 ± 1	3.3 ± 1%	3.4 ± 0.3	
837 ± 2	$2 < T_{\frac{1}{2}} < 4$	0.54 ± 0.1	Not mentioned in previou literature
657 ± 1	3.3 ± 1%	29 ± 3	
589 ± 1	3.3 ± 1%	3.3 ± 0.3	
528 ± 2	$2 < T_{\frac{1}{2}} < 5$	0.7 ± 0.7	Not mentioned in previou literature
374 ± 1	3.3 ± 1%	5.5 ± 1	
283 ± 1	3.3 ± 1%	44 ± 5	
64 ± 4	3.3 ± 1%	14	Taken from previous work. Our efficiency calib. not good for this energy

* The half life of Cu^{61} was assumed to be 3.3 hr.

CONCLUSIONS

This thesis is a report on work done on the gamma ray spectra of Sn^{113}, Zn^{62}, Cu^{62} and Cu^{61} from May 1965 to May 1966. These studies have been carried out with both NaI(Tl) detectors and a lithium drifted germanium detector.

In the case of Sn^{113} a gamma ray continuum having an end - point of 680 $\pm$ 10 keV has been found using the lithium drifted germanium detector. An upper limit of 0.04% has been set on the 648 keV cross - over transition in In^{113}.

Various reaction products produced by proton bombardment of copper have been studied. In particular the decay scheme of Zn^{62} has been found to be different from that given by Brun et al (Brun et al, 1957). Two new gamma ray transitions at 528 and 837 keV have been found to occur following the decay of Cu^{61}. The work on Cu^{62} proved rather difficult due to the intense positron annihilation radiation present. For this reason our conclusions with respect to this isotope are less definite. Tables 3 and 4 give a list of the gamma rays found in the decay of the cyclotron - produced isotopes studied.

The high resolution of the lithium drifted germanium detector was of particular value throughout this study. Because

of this high resolution it happened only rarely that two peaks were not resolved. This made possible work on sources containing relatively large amounts of impurities since these impurities could be identified and the effects due to their presence corrected for. The small depletion depth of our germanium detector made coincidence work impossible and this made the determination of the decay sequence of the gamma rays difficult in some cases. Some coincidence work was done with NaI(Tl) detectors but their relatively poor resolution made work with them difficult. However the accurate energy determination of the gamma rays, in combination with NaI(Tl) coincidence data, makes it improbable that any decay scheme radically different from that proposed (Figures 8 and 10) could be set up to fit the existing data.

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